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(54) ADHERENT CURABLE COMPOSITION

(57)Abstract:
PROBLEM TO BE SOLVED: To obtain a composition providing a low-viscosity sealing composition, adhesive composition or coating composition without deterioration of their properties by employing as a main component a vinyl polymer containing a cross-linkable silyl group.
SOLUTION: A vinyl polymer containing a cross-linkable silyl group represented by the formula: [Si(R1)2-b(Y)bO]m-Si(R2)3-a(Y)a is employed. In the formula, R1 and R2 are each 1-20C alkyl, 6-20 aryl, 7-20C aralkyl or (R')3SiO- (wherein R' is 1-20C hydrocarbon); Y is hydroxyl or a hydrolyzable group; a is 0-3; b is 0-2; m is 0-19; and a+mb≥1. The vinyl polymer containing a cross-linkable silyl group is preferably a (meth)acrylate polymer prepared using 40 wt.% or more of a (meth)acrylic acid-based monomer. Preferably, the molecular weight distribution is narrow and the weight average molecular weight/ number average molecular weight ratio is less 1.8.

* NOTICES *

CLAIMS

[Claim(s)]
[Claim 1]An adhesive hardenability constituent which uses as the main ingredients a vinyl system polymer which has at least one crosslinkable silyl groups shown by a general formula (1).

$-[Si(R^1)_{2-b}(Y)_bO]_m-Si(R^2)_{3-a}(Y)_a$ (1)

{Each of R^1 and R^2 among a formula An alkyl group of the carbon numbers 1-20, An aryl group of the carbon numbers 6-20, an aralkyl group of the carbon numbers 7-20, or $(R')_3SiO$ - (R' is a univalent hydrocarbon group of the carbon numbers 1-20, and) three R' being the same and differing -- **** -- when the Tori ORGANO siloxy group shown is shown and R^1 or two or more R^2 exist, they may be the same and may differ. When Y shows a hydroxyl group or a hydrolytic basis and two or more Y exists, they may be the same and may differ. a shows 0, 1, 2, or 3, and b shows 0, 1, or 2. m is an integer of 0-19. However, it shall satisfy that it is a+mb>=1. }
[Claim 2]The adhesive hardenability constituent according to claim 1 whose ratio of weight average molecular weight and a number average molecular weight which were measured with gel permeation chromatography of a vinyl system polymer is less than 1.8.
[Claim 3]The adhesive hardenability constituent according to claim 1 or 2 whose vinyl system polymer is an acrylic (meta) polymer.
[Claim 4]It is an adhesive hardenability constituent given in any 1 paragraph among claims 1-3, wherein a manufacturing method of a vinyl system polymer is a living-radical-polymerization method.
[Claim 5]It is an adhesive hardenability constituent given in any 1 paragraph among claims 1-4 a manufacturing method of a vinyl system polymer using an organic halogenated compound or a sulfonyl halide compound as an initiator, making a transition metal complex a catalyst, and carrying out the radical polymerization of the vinyl system monomer.
[Claim 6]It is an adhesive hardenability constituent given in any 1 paragraph among claims 1-5 which have the crosslinkable silyl groups shown by a general formula (1) in at least one molecular chain terminals.
[Claim 7]A process:(1) organic halogenated compound of the following [polymer / which has at least one crosslinkable silyl groups shown by a general formula (1) / vinyl system], Or a vinyl system polymer which has halogen at the end by using a sulfonyl halide compound as an initiator and carrying out the radical polymerization of the vinyl system monomer by making a transition metal complex into a catalyst is manufactured, (2) By making a oxy anion which has an alkenyl group react, and replacing halogen, It is an adhesive hardenability constituent given in any 1 paragraph among claims 1-6 which are the polymers obtained by; to which a hydrosilane compound which has the crosslinkable silyl groups which manufactures a vinyl system polymer which has an alkenyl group at the end, and is shown by (3) general formulas (1) is made to react.
[Claim 8]By polymerizing by a living-radical-polymerization method, a vinyl system polymer which has the crosslinkable silyl groups shown by a general formula (1) the following process:(1) vinyl system monomers, Manufacture a vinyl system polymer and a vinyl system polymer which has an alkenyl group at the end by making a compound which (2) Continues and has at least two low alkenyl groups of polymerization nature react is manufactured, It is an adhesive hardenability constituent given in any 1 paragraph among claims 1-6 which are the polymers obtained by; changed into a silyl group content substituent to which a hydrosilane compound which has the crosslinkable silyl groups which shows an end alkenyl group by the general formula 1 is made to react.

[Claim 9]It is an adhesive hardenability constituent given in any 1 paragraph among claims 1-8 containing a compound which has an organic group which has at least one of nitrogen, oxygen, and sulfur atoms in a monad as an adhesion promoter, and crosslinkable silyl groups.
[Claim 10]An adhesive hardenability constituent is a constituent given in any 1 paragraph among claims 1-9 which are sealing material compositions.
[Claim 11]The sealing material composition according to claim 10 packed up by 1 liquid type so that bridge construction hardening could be carried out by absorbing hygroscopic surface moisture.
[Claim 12]An adhesive hardenability constituent is a constituent given in any 1 paragraph among claims 1-9 which are pressure sensitive adhesive compositions.
[Claim 13]The pressure sensitive adhesive composition according to claim 12 containing a tackifier.
[Claim 14]An adhesive hardenability constituent is a constituent given in any 1 paragraph among claims 1-9 which are the constituents for paints.
[Claim 15]An adhesive hardenability constituent is a constituent given in any 1 paragraph among claims 1-9 which are the constituents for powder coatings.

[Translation done.]

DETAILED DESCRIPTION

[Detailed Description of the Invention]
[0001]
[Field of the Invention]This invention relates to the hardenability constituent, i.e., the adhesive hardenability constituent, which have an adhesive property or adhesiveness. Excel in weatherability and heat resistance, in more detail, since viscosity is low, deal with it, and a sex is good, And ingredient[one]-izing is possible and it excels in the sealing material composition excellent also in paintwork and weatherability, and heat resistance, and since viscosity is low, the amount of the solvent used can be reduced remarkably (high-solid-izing), and it is related with the small constituent for paints and pressure sensitive adhesive composition of an environmental impact.
[0002]
[Description of the Prior Art]As a sealing material composition which is excellent in weatherability and heat resistance until now, It has the hydroxyl group or hydrolytic basis combined with the silicon atom, and the sealant of the silicone series containing the silicon content group (henceforth crosslinkable silyl groups) which can construct a bridge by forming a siloxane bond is known well, and is widely used for the building use etc. The theme of the sealing material of a polyisobutylene system which has crosslinkable silyl groups at the end is carried out as a weatherproof sealing material new type these days. Even if it does not add a tackifier, acrylic pressure sensitive adhesive has the adhesion characteristics which balance was able to take, and is produced in large quantities along with the crude rubber system binder. Since cohesive force is insufficient for especially acrylic pressure sensitive adhesive from the problem of a molecular weight and molecular weight distribution, generally this has been improved according to bridge construction. As a crosslinking method, various kinds of forms are developed and For example, a polyisocyanate compound, The method of adding cross linking agents, such as an epoxy compound, polyvalent carboxylic acid, polyamine compound, phenol resin, and sulfur compounds, or the method of making the acrylic polymer which has crosslinkable silyl groups construct a bridge under condensation catalyst existence is proposed. Especially the binder that uses as the main ingredients the acrylic polymer which has crosslinkable silyl groups has the feature of excelling in weatherability, in order to carry out bridge construction hardening by a siloxane bond.
[0003]By carrying out bridge construction hardening under existence of a suitable condensation catalyst, the coat excellent in weatherability is obtained and the vinyl system which has crosslinkable silyl groups in intramolecular, or (meta) the acrylic polymer is used as a high weatherability paint of a solvent system and a drainage system.
[0004]
[Problem(s) to be Solved by the Invention]Although the silicone system sealing compound is excellent in movement-proof nature, low-temperature workability, etc. other than weatherability, that the adhesion of a paint is bad, polluting the masonry joint circumference, etc. are pointed out as a fault. Although the polyisobutylene system sealing material is excellent also in moisture permeability-proof besides weatherability, since viscosity is high and workability cannot let a bad thing and hygroscopic surface moisture pass easily, it is pointed out that ingredient[one]-izing is difficult etc. On the other hand, it is known that weatherability is good and the vinyl system or (meta) the acrylic polymer has the possibility as base polymer for high weatherability sealing materials. Although the vinyl system which has crosslinkable silyl groups as mentioned above, or (meta) the acrylic polymer is put in practical use as a high weatherability paint, it is manufactured by usually carrying out

copolymerization of the acrylic system monomer which has crosslinkable silyl groups (meta), and other vinyl system monomers. For this reason, since the silyl group which is a cross-linking group is introduced at random into the chain, it is rather difficult to use for the elastic sealing compound as which a low modulus quantity extension characteristic is required. When the vinyl system or (meta) the acrylic polymer had crosslinkable silyl groups in the main chain terminal, it was expected to be able to use as a weatherproof sealing material new type, but it was not easy to manufacture such (meta) an acrylic polymer until now. When the binder using the acrylic polymer which has crosslinkable silyl groups (meta) also uses a low molecular weight body in order to hypoviscosity-ize it since the silyl group is introduced at random into the chain, The interval of the point constructing a bridge becomes small and the bridge construction thing produced by making a silyl group construct a bridge has the problem that elastic character required for a binder is not obtained. In order to give elastic character to a binder, this polymer is used as the amount object of polymers, the amount of the silyl group content monomer used which carries out copolymerization is lessened, and there is the method of enlarging the interval of the point constructing a bridge. However, since it will become hyperviscosity or a solid if this polymer is used as the amount object of polymers, in order to use it as a binder, it is necessary to hypoviscosity-ize using quite a lot of solvents. . After carrying out coating of the solvent type binder to substrates, such as a film, it vaporizes a solvent. Since a lot of thermal energies may be consumed to this, and it may become a cause of an outbreak of a fire or it may have an adverse effect on a human body, solvent[non-]-izing or high solid-ization is called for and there is a limit in polymers quantification of a polymer. Using for base polymer of a binder the acrylic copolymer in which the silyl functional group which the molecular weight of an acrylic (meta) polymer is comparatively small in order to solve this problem, and is hypoviscosity enough, and causes bridge construction or chain extension was introduced into the end of the polymer is proposed. However, it was not easy to manufacture until now the acrylic polymer which has crosslinkable silyl groups at the end (meta). These days, the solvent type paint with which a lot of solvents vaporize from the rise of concern to earth environment is kept at arm's length, and much more high solid-ization of the paint is called for. In order to attain high solid-ization of a vinyl system or (meta) acrylic lacquer generally, in order to secure the plasticity at the time of coating, hypoviscosity-izing is required, and it is necessary to make the molecular weight of a polymer small. However, when a molecular weight is made small, there is a problem that the weatherability which a vinyl system or (meta) an acrylic polymer originally has will be lost. As one method of solving this, a vinyl system or (meta) the molecular weight distribution of an acrylic polymer, That is, the ratio (Mw/Mn) of the weight average molecular weight (Mw) and the number average molecular weight (Mn) which were measured with gel permeation chromatography is made small, and how to attain high solid-ization can be considered by lowering the viscosity of a polymer. However, the vinyl system or (meta) the acrylic polymer used for a paint is usually manufactured by free radical polymerization, and only the large (2 or more [Usually]) polymer of molecular weight distribution is obtained. It was not easy to obtain the narrow vinyl system or (meta) the acrylic polymer of molecular weight distribution until now. If a molecular weight is made small for hypoviscosity-izing, when the interval of the point constructing a bridge becomes short and hardens in connection with it, it will become a coat with dramatically high crosslinking density. As a result, the problem that the coat obtained is very deficient in elastic character, and it cannot follow in footsteps of modification [a coated object] is produced. One means to solve this problem is using the vinyl system which has a silyl group which is a cross-linking functional group at the end of a main chain, or (meta) an acrylic polymer as the main ingredients of a paint. Maintaining a molecular weight at suitable length by having crosslinkable silyl groups at the end, distance between the points constructing a bridge can be effectively made large, and, as a result, elastic character can be given to the coat after hardening. However, it was not easy to manufacture until now the vinyl system which has crosslinkable silyl groups at the end, or (meta) an acrylic polymer. In order to obtain the sealing material composition which needs the hardened material of high elongation, a pressure sensitive adhesive composition, and a coating composition, As a trial which compounds the acrylic polymer which has crosslinkable silyl groups at the end (meta), for example, In JP,3-14068,B, an acrylic system monomer (meta) Crosslinkable-silyl-groups content mercaptan, In [again] JP,4-55444,B the method of polymerizing under existence of the disulfide which has crosslinkable silyl groups, and the radical polymerization initiator which has crosslinkable silyl groups, The method of polymerizing an acrylic system monomer under existence of a

crosslinkable-silyl-groups content hydrosilane compound or a tetra halo silane is indicated. However, it is difficult to introduce crosslinkable silyl groups into both ends certainly, and these methods are insufficient in the field of hardenability -- a gel fraction is low. By using hydroxyl group content polysulfide for JP,6-211922,A in large quantities to an initiator, First, the acrylic polymer which has a hydroxyl group at the end is compounded, and the room-temperature-curing nature constituent using the acrylic polymer which has crosslinkable silyl groups at the end obtained by changing a hydroxyl group further (meta) is indicated. Although the acrylic polymer which has crosslinkable silyl groups by a comparatively high ratio at the end in this method is obtained, hydroxyl group content polysulfide which is an expensive chain transfer agent must be used in large quantities, and there is a manufacturing process top problem. Since molecular weight distribution becomes large, there is also a problem that the viscosity of a polymer becomes high. Namely, in order to obtain an adhesive hypoviscosity hardenability constituent, a sealing material composition, a pressure sensitive adhesive composition, and a coating composition, without reducing physical properties, it is required to obtain the narrow vinyl system or (meta) the acrylic polymer of molecular weight distribution which has crosslinkable silyl groups in an end or a main chain, but. It was not easy to manufacture until now the acrylic polymer which has crosslinkable silyl groups at the end (meta).

[0005]

[Means for Solving the Problem]By using a vinyl system polymar which has at least one crosslinkable silyl groups as the main ingrediants of an adhesive hardenability constituent, it found out solving an aforementioned problem and this invention was reached.

[0006]

[Embodiment of the Invention]This invention is an adhesive hardenability constituent using the vinyl system polymer which has at least one crosslinkable silyl groups shown by a general formula (1) as the main ingredients.

$-[Si(R^1)_{2-b}(Y)_bO]_m-Si(R^2)_{3-a}(Y)_a$ (1)

{Each of R^1 and R^2 among a formula The alkyl group of the carbon numbers 1-20, The aryl group of the carbon numbers 6-20, the aralkyl group of the carbon numbers 7-20, or $(R')_3SiO-$ (R' is a univalent hydrocarbon group of the carbon numbers 1-20, and) three R' being the same and differing -- **** -- when the Tori ORGANO siloxy group shown is shown and R^1 or two or more R^2 exist, they may be the same and may differ. When Y shows a hydroxyl group or a hydrolytic basis and two or more Y exists, they may be the same and may differ. a shows 0, 1, 2, or 3, and b shows 0, 1, or 2. m is an integer of 0-19. However, it shall satisfy that it is a+mb>=1. }

Especially as a hydrolytic basis shown by the above-mentioned Y, it is not limited, but can use a publicly known thing conventionally, and specifically, The point of hydrogen, a halogen atom, an alkoxy group, an acyloxy group, a KETOKISHI mate group, an amino group, an amide group, an aminooxy group, a sulfhydryl group, an alkanyloxy group, etc. being mentioned, and hydrolysis nature being mild and being easy to deal with it to especially an alkoxy group is preferred. This hydrolytic basis and a hydroxyl group can be combined with one silicon atom in the 1-3 ranges, and total of a+mb, i.e., a hydrolytic basis, has the preferred range of 1-5. When a hydrolytic basis and a hydroxyl group join together in [two or more] a cross-linking silicon group, they may be the same or may differ. The number of the silicon atoms which constitute a cross-linking silicon compound may be one, and although it may be two or more pieces, there may be a case of the silicon atom connected by the siloxane bond to about 20 pieces.

[0007]As a monomer which constitutes the main chain of the vinyl system polymer which has at least one crosslinkable silyl groups of a general formula (1), there are no restrictions in particular and various kinds of things can be used. Especially as a vinyl system monomer used for manufacture of the main chain of the vinyl system polymer of this invention, it is not limited but various kinds of things can be used. If it illustrates, acrylic acid (meta), methyl acrylate (meta), (Meta) Ethyl acrylate, acrylic acid (meta)-n-propyl, acrylic acid (meta) isopropyl, (Meta) Acrylic acid-n-n-butyl, isobutyl acrylate (meta), (Meta) Acrylic acid-tert-butyl, acrylic acid (meta)-n-pentyl, (Meta) Acrylic acid-n-haxyl, acrylic acid (meta) cyclohexyl, (Meta) Acrylic acid-n-heptyl, acrylic acid (meta)-n-octyl, (Meta) Acrylic acid-2-ethylhexyl, acrylic acid (meta) nonyl, (Meta) Decyl acrylate, acrylic acid (meta) dodecyl, acrylic acid (meta) phanyl, (Meta) Acrylic acid toluyl, acrylic acid (mata) benzyl, acrylic acid (meta)-2-methoxy ethyl, (Meta) Acrylic acid-3-methoxy propyl, acrylic acid (meta)-2-hydroxyethyl,

(Meta) Acrylic acid-2-hydroxypropyl, acrylic acid (meta) stearyl, metaglycidyl acrylate (meta), acrylic acid (meta) 2-aminoethyl, gamma-(methacryloyl oxypropyl) trimethoxysilane, the ethyleneoxide addition of acrylic acid (meta), (Meta) Acrylic acid trifluoromethyl methyl, an acrylic acid (meta) 2-trifluoro methylethyl, (Meta) Acrylic acid-2-perfluoro athylathyl, acrylic acid (meta) 2-perfluoro ethyl-2-perfluoro butylethyl, (Meta) Acrylic acid 2-perfluoro ethyl, acrylic acid (meta) perfluoro methyl, (Meta) Acrylic acid JIPA fluoromethylmethyl, acrylic acid (meta) 2-perfluoro methyl-2-perfluoro ethylmethyl, (Meta) Acrylic acid 2-perfluoro hexylethyl, acrylic acid (meta) 2-perfluoro decylethyl, (Meta) Acrylic acid series (meta) monomers, such as acrylic acid 2-perfluoro hexadecylethyl; Styrene, Styrene system monomers, such as vinyltoluene, alpha-methylstyrene, KURORU styrene, styrene sulfonic acid, and its salt; Perfluoro ethylene, Monoalkyl ester and dialkyl ester of silicon content vinyl system monomer; maleic anhydrides, such as fluoride content vinyl monomer; vinyltrimetoxysilane, such as perfluoro propylene and vinylidene fluoride, and vinyltriethoxysilane, maleic acid, and maleic acid; Fumaric acid, Monoalkyl ester and dialkyl ester of fumaric acid; Maleimide, Methylmaleimide, ethylmaleimide, propyl maleimide, butylmaleimide, Hexylmaleimide, octylmaleimide, dodecylmaleimide, stearyl maleimide, Maleimide system monomers, such as phenylmaleimide and cyclohexylmaleimide; Acrylonitrile, Nitrile group content vinyl system monomers, such as a methacrylonitrile; Acrylamide, Amide group content vinyl system monomers, such as methacrylamide; Vinyl acetate, Conjugated dienes, such as alkenes; butadiene, such as vinyl ester; ethylene, such as vinyl propionate, vinyl pivalate, benzoic acid vinyl, and vinyl cinnamic acid, and propylene, and isoprene; VCM/PVC, a vinylidene chloride, an allyl chloride, allyl alcohol, etc. are mentioned. These may be used independently, and even if it carries out copolymerization of the plurality, they are not cared about. Acrylic acid (meta) expresses acrylic acid and/, or mathacrylic acid with the abova-mentioned expressive form.

[0008]The acrylic (meta) polymer obtained by compounding the crosslinkable silyl groups of a general formula (1) in the above-mentioned monomer with the vinyl system polymer which it has at least one piece, using an acrylic acid series (meta) monomer 40% of the weight or more is more preferred from a physical-properties side. Although there is no restriction in particular about the molecular weight of the vinyl system polymer which has at least one crosslinkable silyl groups, it is preferred that it is in the range of 500-100000. The original characteristic of a vinyl system polymer is it hard to be revealed that a molecular weight is 500 or less, and handling becomes it difficult that it is 100000 or more.

[0009]There is no restriction in particular about the ratio (Mw/Mn) of the weight average molecular weight (Mw) and the number average molecular weight (Mn) which measured crosslinkable silyl groups according to the molecular weight distribution of the vinyl system polymer which it has at least one piece, i.e., gel permeation chromatography. However, in order easily and to acquire hardened material physical properties sufficient moreover, the molecular weight distribution of narrow one is preferred. [by stopping low the viscosity at the time of considering it as an adhesiva herdenability constituant] It is 1.3 or less still more preferebly 1.4 or lesa still more preferably 1.5 or less still more preferably 1.6 or lesa still more preferably 1.7 or less more preferably [as a value of molecular weight distribution, less than 1.8 are preferred, and].

[0010]The vinyl system polymer which has at least one crosslinkable silyl groups can be obtained by the various polymerizing methods, and the method in particular is not limited. By however, the point of the flexibility of a monomer, and the ease of control to a radical polymerization method, The vinyl system polymer which has a specific functional group which can introduce crosslinkable silyl groups directly or can be changed into crosslinkable silyl groups at one step or several steps of reactions is obtained, The method of obtaining the vinyl system polymer which has at least one crosslinkable silyl groups is more preferred by changing this specific functional group into crosslinkable silyl groups.

[0011]The radical polymerization method used in the method of compounding the vinyl system polymer which has a specific functional group containing crosslinkable silyl groups, It can classify into the "control radical polymerization method" which can introduce a specific functional group into "the general radical polymerization method" to which copolymerization of the monomer which has a specific functional group, and the vinyl system monomar is only carried out, and to carry out, and the position by which the end etc. were controllad using an azo compound, a peroxide, etc. as a polymerization initiator. "A general radical polymerization method" is a simple method, and it is [method] usable also in this invantion. However, since the monomer which has a specific functional

group in this method is not introduced into a probable polymer, when it is going to obtain a polymer with a high rate of organic-functiona-izing. It is necessary to use this monomer quite in large quantities, and there is a problem that the rate of a polymer that this specific functional group is not introduced becomes large, by use in small quantities conversely. Since it is a free radical polymerization, the problem that only a large polymer with high viscosity is obtained also has molecular weight distribution. A "control radical polymerization method" further, By polymerizing using the chain transfer agent which has a specific functional group. It can classify into the "living-radical-polymerization method" the polymer of the molecular weight as a design is obtained mostly, by growing without the "chain transfer agent method" the vinyl system polymer which has a functional group at the end is obtained, and a polymerization growth end causing a termination reaction etc.

[0012]Although the "chain transfer agent method" can obtain a polymer with a high rate of organic-functions-izing and it is usable also in this invention, the chain transfer agent which has quite a lot of specific functional groups to an initiator is required, and there is a problem on the financial side also including processing. Like the above-mentioned "general radical polymerization method", since it is a free radical polymerization, the problem that only the polymer whose viscosity it is large and is high is obtained also has molecular weight distribution.

[0013]Unlike these polymerizing methods, a "living-radical-polymerization method". Since a rate of polymerization is high, and the termination reaction by radical coupling etc. occurs easily, though it is a radical polymerization made difficult [control], While a termination reaction does not occur easily and the narrow polymer (Mw/Mn is 1.1 to about 1.5) of molecular weight distribution is obtained, a molecular weight is freely controllable by the preparation ratio of a monomer and an initiator.

[0014]Therefore, since the monomer which can obtain a polymer with molecular weight distribution narrow [a "living-radical-polymerization method"] and low viscosity, and also has a specific functional group can be introduced into the almost arbitrary positions of a polymer, As a manufacturing method of the vinyl system polymer which has the above-mentioned specific functional group, it is more desirable. Although living polymerization means the polymerization in which an end always continues having activity and the chain grows in the narrow sense, the pseudo-living polymerization which grows while that by which the end was inactivated, and the activated thing are generally in an equilibrium situation is also included. The definition in this invention is also the latter.

[0015]As for the "living-radical-polymerization method", research is positively made into various groups in recent years. As the example, for example A journal OBU American chemical society (J. Am.Chem.Soc.), 1994, 116 volumes, the thing using a cobalt porphyrin complex as shown in 7943 pages, Macro leakage-at-bulb KYURUZU (Macromolecules), 1994, The thing using radical scavengers, such as a nitroxide compound as shown in 27 volumes and 7228 pages, The "atom transfer radical polymerization" (Atom Transfer Radical Polymerization:ATRP) etc. which uses an organic halogenated compound etc. as an initiator and makes a transition metal complex a catalyst is raised.

[0016]Also in a "living-radical-polymerization method", an organic halogenated compound or a sulfonyl halide compound An initiator, The "atom-transfer-radical-polymerization method" which polymerizes a vinyl system monomer by making a transition metal complex into a catalyst, As a manufacturing method of the vinyl system polymer which in addition to the feature of the above-mentioned "living-radical-polymerization method" has halogen comparatively advantageous to a functional group conversion reaction etc. at the end, and has a specific functional group from the flexibility of a design of an initiator or a catalyst being large, it is still more desirable. As this atom-transfer-radical-polymerization method, Matyjaszewski et al. [for example,], Journal OBU American chemical society (J. Am.Chem.Soc.) 1995, 117 volumes, 5614 pages, macro leakage-at-bulb KYURUZU (Macromolecules) 1995, 28 volumes, 7901 pages, science (Science) 1996, 272 volumes, 28 volumes, 1721 pages, etc. will be mentioned in 866 pages, WO 96/No. 30421 gazette, WO 97/No. 18247 gazette or Sawamoto et al., and macro leakage-at-bulb KYURUZU (Macromolecules) 1995.

[0017]The organic halogenated compound which has an organic halogenated compound, especially a reactant high carbon-halogen bond in this atom transfer radical polymerization. (For example, the carbonyl compound which has halogen in an alpha position and the compound which has halogen in a benzylic position), or a sulfonyl halide compound is used as an initiator. As a transition metal complex used as a catalyst of the above-mentioned atom transfer radical polymerization, the complex used as

a central metal can use the 7th fellows of the periodic table, eight fellows, nine fellows, ten fellows, and 11 group elements. As a desirable thing, the complex of zerovalent copper, univalent copper, a divalent ruthenium, divalent iron, or divalent nickel is mentioned. Especially, a copper complex is preferred. If a univalent copper compound is illustrated concretely, they will be a cuprous chloride, the first copper of bromination, a cuprous iodide, a cuprous cyanide, copper I oxide, the first copper of perchloric acid, etc. When using a copper compound, in order to improve catalytic activity 2,2'-bipyridyl and its derivative. Ligands, such as polyamine, such as a 1,10-phenanthroline and its derivative, tetramethylethylenediamine, pentamethyl diethylenetriamine, and hexamethyl tris(2-aminoethyl) amine, are added. The tristriphenyl phosphine complex ($RuCl_2(PPh_3)_3$) of divalent ruthenium chloride is also preferred as a catalyst. When using a ruthenium compound as a catalyst, aluminum alkoxides are added as an activator. The screw triphenyl phosphine complex of divalent iron ($FeCl_2(PPh_3)_2$), The screw triphenyl phosphine complex ($NiCl_2(PPh_3)_2$) of divalent nickel and the screw tributyl phosphine complex ($NiBr_2(PBu_3)_2$) of divalent nickel are also preferred as a catalyst.

[0018]As a monomer of a vinyl system used in this polymerization, there are no restrictions in particular and all the already illustrated things can be used conveniently. The above-mentioned polymerization reaction can be performed in a non-solvent or various kinds of solvents. As a solvent, for example Hydrocarbon system solvent; diethylether, such as benzene and toluene, Ether system solvents, such as a tetrahydrofuran; Halogenated hydrocarbon system solvent; acetone, such as a methylene chloride and chloroform, Ketone solvent, such as methyl ethyl ketone and methyl isobutyl ketone; Methanol, Ethanol, propanol, isopropanol, n-butyl alcohol, Alcoholic solvent, such as tert-butyl alcohol; carbonate system solvents, such as ester solvent; ethylene carbonate, such as nitrile system solvent; ethyl acetate, such as acetonitrile, propionitrile, and benzonitrile, and butyl acetate, and propylene carbonate, etc. are mentioned. These can be independent, or can mix two or more sorts, and can be used. The above-mentioned polymerization can be performed in 0-200 **, and it is the range of room temperature -150 ** preferably.

[0019]Although the vinyl system polymer which has at least one crosslinkable silyl groups shown by the general formula 1 can be obtained using the method of illustrating below, it is not necessarily limited to these. As a synthesizing method of the vinyl system polymer which it has at least one piece, crosslinkable silyl groups, (A) The hydrosilane compound which has crosslinkable silyl groups in the vinyl system polymer which has at least one alkenyl group, By the method (C) radical polymerization which makes the compound which has in a monad a basis which can react to crosslinkable silyl groups and a hydroxyl group like an isocyanate group react to the vinyl system polymer which has at least one method (B) hydroxyl group made to add under hydrosilylation catalyst existence, When compounding a vinyl system polymer by the method (D) radical polymerization to which the compound it has a compound, and the alkenyl group and crosslinkable silyl groups of polymerization nature in one molecule is made to react when compounding a vinyl system polymer, Method; etc. to which the compound which has stable carbanion with crosslinkable silyl groups in one molecule in the vinyl system polymer which has at least one high carbon-halogen bond of method (E) reactivity which uses the chain transfer agent which has crosslinkable silyl groups is made to react are raised.

[0020]The vinyl system polymer which has at least one alkenyl group used by the method of (A) is obtained by various methods.Although a synthesizing method is illustrated below, it is not necessarily limited to these.

(A-a) A method to which the compound it has a compound and an alkenyl group of polymerization nature and a low alkenyl group of polymerization nature in a monad which is mentioned, for example to the following general formula (2) when compounding a vinyl system polymer by a radical polymerization is made to react as the 2nd monomer.

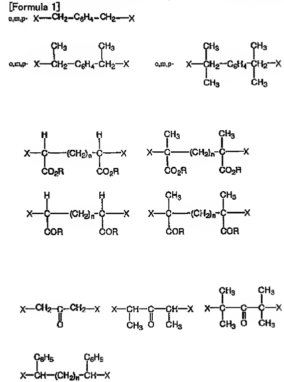
$H_2C=C(R^3)-R^4-R^5-C(R^6)=CH_2$ (2)

(R^3 shows hydrogen or a methyl group among a formula, R^4 shows -C(O) O- or o-, m-, and p-phenylene group, and R^5 may show the divalent organic group of direct coupling or the carbon numbers 1-20, and may include one or more ether bonds.) R^6 shows hydrogen or the alkyl group of the carbon numbers 1-10, the aryl group of the carbon numbers 6-10, or the aralkyl group of the carbon numbers 7-10.

Although there is no restriction at the stage to which the compound it has a compound and an alkenyl group of polymerization nature and a low alkenyl group of polymerization nature in a monad is made to react, it is especially living radical polymerization, When it expects rubber character, it is preferred to make it react as the 2nd monomer after the telophase of a polymerization reaction or the ending reaction of a predetermined monomer.
[0021](A-b) When compounding a vinyl system polymer by living radical polymerization, A method to which the compound which has at least two low alkenyl groups of polymerization nature, such as 1,5-hexadiene, 1,7-octadien, and 1,9-decadiene, for example is made to react after the telophase of a polymerization reaction, or the ending reaction of a predetermined monomer.
(A-c) How to make various kinds of organic metallic compounds which have an alkenyl group like organic tin, such as allyl tributyl tin and allyl trioctyl tin, for example react to the vinyl system polymer which has at least one reactant high carbon-halogen bond, and to replace halogen by it.
[0022](A-d) How to make the stabilization carbanion which has an alkenyl group which is mentioned to a general formula (3) react to the vinyl system polymer which has at least one reactant high carbon-halogen bond, and to replace halogen by it.
$M^+C^-(R^7)(R^8)-R^9-C(R^6)=CH_2$ (3)
inside of formula, and R^6 — the above — the same — both R^7 and R^8 are electron withdrawing groups which stabilize carbanion C^-, or, in one side, another side shows hydrogen, the alkyl group of the carbon numbers 1-10, or a phenyl group by said electron withdrawing group. R^9 may show the divalent organic group of direct coupling or the carbon numbers 1-10, and may include one or more ether bonds. M^+ shows alkali metal ion or the 4th class ammonium ion.
As an electron withdrawing group of R^7 and R^8, especially the thing that has the structure of $-CO_2R$, $-C(O)R$, and $-CN$ is preferred.
[0023](A-e) To the vinyl system polymer which has at least one reactant high carbon-halogen bond. For example, make the metal simple substance or organic metallic compound like zinc act, and a eno rate anion is prepared, How to make after an appropriate time react to the electrophilicity compound which has alkenyl groups, such as an alkenyl group content compound which has a leaving group like halogen or an acetyl group, a carbonyl compound which has an alkenyl group, an isocyanate compound which has an alkenyl group, and acid halide which has an alkenyl group.
[0024](A-f) How to make the oxy anion or carboxylate anion which has an alkenyl group as shown, for example in the general formula (4) or (5) react to the vinyl system polymer which has at least one reactant high carbon-halogen bond, and to replace halogen by it.
$H_2C=C(R^6)-R^{10}-O^-M^+$ (4)
(R^6 and M^+ are the same as the above among a formula.) R^{10} may include one or more ether bonds by the divalent organic group of the carbon numbers 1-20.
$H_2C=C(R^6)-R^{11}-C(O)O^-M^+$ (5)
(R^6 and M^+ are the same as the above among a formula.) It is mentioned that R^{11} may include one or more ether bonds by the divalent organic group of direct coupling or the carbon numbers 1-20 etc.
[0025]The synthetic method of the vinyl system polymer which has at least one above-mentioned reactant high carbon-halogen bond as an example, (E-a) a carbon tetrachloride as shown in JP,4-132706,A in a radical polymerization, ethylene chloride, carbon tetrabromide, and a methylene bromide — how (chain transfer agent method) to use a halogenide [like] for a chain transfer agent.
[0026](E-b) The above organic halogenated compounds etc. are used as an initiator, and although atom-transfer-radical-polymerization method; etc. which make a transition metal complex a catalyst are mentioned, it is not necessarily limited to these. Although the vinyl system polymer which has at least one alkenyl group is possible also for obtaining from the vinyl system polymer which has at least one hydroxyl group and can use the method of illustrating below, it is not necessarily limited to these. How to make a base like sodium methoxide (A-g) act on the hydroxyl group of the vinyl system polymer which has at least one hydroxyl group, and make it react to an alkenyl group content halogenide like an allyl chloride.
[0027](A-h) A method to which alkenyl group content isocyanate compounds, such as an allylisocyanate, are made to react.

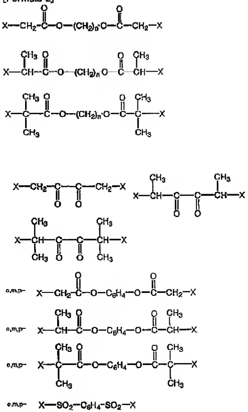
(A-i) A method (meta) to which alkenyl group content acid halide like acrylic acidchloride is made to react under base existence, such as pyridine.
(A-j) Method; etc. which make alkenyl group content carboxylic acid, such as acrylic acid, react under existence of an acid catalyst are mentioned.
[0028]When halogen does not participate in the method of introducing an alkenyl group like this invention (A-a) (A-b) directly, it is preferred to compound a vinyl system polymer using a living-radical-polymerization method. The method of of the point that control is easier to (A-b) is still more preferred. When introducing an alkenyl group by changing halogen of the vinyl system polymer which has at least one reactant high carbon-halogen bond, The organic halogenated compound which has at least one reactant high carbon-halogen bond, Or it is preferred to use the vinyl system polymer which is obtained by using a sulfonyl halide compound as an initiator and carrying out the radical polymerization of the vinyl system monomer by making a transition metal complex into a catalyst (atom-transfer-radical-polymerization method) and which has at least one reactant high carbon-halogen bond at the end. The method of of the point that control is easier to (A-f) is still more preferred.
[0029]Although there is no restriction in particular as a hydrosilane compound which has crosslinkable silyl groups, if a typical thing is shown, the compound shown by a general formula (6) will be illustrated.
$H-[Si(R^1)_{2-b}(Y)_bO]_m-Si(R^2)_{3-a}(Y)_a$ (6)
(R^1, R^2, a, b, m, and Y are the same as the above among a formula.)
Also in these hydrosilane compound, it is especially a general formula (7).
$H-Si(R^2)_{3-a}(Y)_a$ (7)
the compound which has a cross-linking group shown by (the inside of a formula, R^2, Y, and a are the same as the above) — acquisition — it is desirable from an easy point.
[0030]When making the hydrosilane compound which has the above-mentioned crosslinkable silyl groups add to an alkenyl group, a transition metal catalyst is usually used. The thing which, for example, made carriers, such as a platinum simple substance, alumina, silica, and carbon black, distribute a platinum solid as a transition metal catalyst, A complex with chloroplatinic acid, chloroplatinic acid, alcohol, aldehyde, ketone, etc., a platinum-olefin complex, and a platinum (0)-divinyl tetramethyl disiloxane complex are mentioned. As an example of catalysts other than a platinum compound, $RhCl(PPh_3)_3$, $RhCl_3$, $RuCl_3$, $IrCl_3$, $FeCl_3$, $AlCl_3$, $PdCl_2-H_2O$, $NiCl_2$, $TiCl_4$, etc. are mentioned.
[0031](B) And the manufacturing method of the vinyl system polymer which has at least one hydroxyl group used by the method of (A-g) - (A-j) is not limited to these methods, although the following methods are illustrated.
(B-a) A method to which the compound it has a compound, and the alkenyl group and hydroxyl group of polymerization nature in a monad which is mentioned, for example to the following general formula (8) when compounding a vinyl system polymer by a radical polymerization is made to react as the 2nd monomer.
$H_2C=C(R^3)-R^4-R^5-OH$ (8)
(The inside of a formula, R^3, R^4, and R^5 are the same as the above)
Although there is no restriction at the stage to which the compound it has a compound, and the alkenyl group and hydroxyl group of polymerization nature in a monad is made to react, it is especially living radical polymerization, and when it expects rubber character, it is preferred to make it react as the 2nd monomer after the telophase of a polymerization reaction or the ending reaction of a predetermined monomer.
[0032](B-b) A method to which 10-UNDESE Norian, 5-hexenol, and alkenyl alcohol like allyl alcohol are made to react after the telophase of a polymerization reaction, or the ending reaction of a predetermined monomer for example when compounding a vinyl system polymer by living radical polymerization.
(B-c) For example, the method of carrying out the radical polymerization of the vinyl system monomer, using so much a hydroxyl group content chain transfer agent like the hydroxyl group content polysulfide shown in JP,5-262808,A.

(B-d) For example, the method of carrying out the radical polymerization of the vinyl system monomer using hydrogen peroxide or a hydroxyl group content initiator as shown in JP,6-239912,A and JP,8-283310,A.
(B-a) For example, tha method of carrying out the radical polymerization of the vinyl system monomer, using alcohols as shown in JP,6-116312,A superfluously.
(B-f) For axample, the method of introducing a hydroxyl group into an end by making halogen of the vinyl system polymer which has a reactant high carbon-halogen bond in at least one piece react to hydrolysis or hydroxyl group-containing compound by a method as shown in JP,4-132706,A etc.
(B-g) How to make the stabilization carbanion which has a hydroxyl group which is mentioned to a general formula (9) react to the vinyl system polymer which has at least one reactant high carbon-halogen bond, and to replace halogen by it.
$M^+C^-(R^7)(R^8)-R^9-OH$ (9)
(It is the same as the inside of a formula, and R^7, R^8, R^9 and ******)
As an electron withdrawing group of R^7 and R^8, especially the thing that has the structure of $-CO_2R$, $-C(O)R$, and $-CN$ is preferred.
[0033](B-h) How to, make the metal simple substance or organic metallic compound like zinc act on the vinyl system polymer which has at least one reactant high carbon-halogen bond for example, to prepare a eno rate anion, and to make aldehyde or ketone react to after an appropriate time.
(B-i) How to make the oxy anion or carboxylate anion which has a hydroxyl group as shown, for example in the general formula (10) or (11) react to the vinyl system polymer which has at least one reactant high carbon-halogen bond, and to replace halogen by it.
$HO-R^{10}-O^-M^+$ (10)
(The inside of a formula, R^{10}, and M^+ are the same as the above)
$HO-R^{11}-C(O)O^-M^+$ (11)
(The inside of a formula, R^{11}, and M^+ are the same as the above)
** is mentioned.
[0034]When halogen does not participate in the method of introducing a hydroxyl group like (B-a) - (B-e) in this invention directly, it is preferred to compound a vinyl system polymer using a living-radical-polymerization method. The method of of the point that control is easier to (B-b) is still more preferred. When introducing a hydroxyl group by changing halogen of the vinyl system polymer which has at least one reactant high carbon-halogen bond, . Obtain by using an organic halogenated compound or a sulfonyl halide compound as an initiator, and carrying out the radical polymerization of the vinyl system monomer by making a transition matal complex into a catalyst (atom-transfer-radical-polymerization method). It is preferred to use for an end the vinyl system polymer which has at least one reactant high carbon-halogen bond. The method of of the point that control is easier to (B-i) is still more prefarred.
[0035]As a compound which has in a monad a basis which can react to crosslinkable silyl groups and a hydroxyl group like an isocyanate group, For example, gamma-isocyanate propyltrimethoxysilana, gamma-isocyanate propylmethyl dimethoxysilane, gamma-isocyanate propyl triethoxysilane, etc. are mentioned, and the catalyst of a urethane-ized reaction generally known as occasion demands can be used.
[0036]As a compound it has a compound, and the alkenyl group and crosslinkable silyl groups of polymerization nature in the monad used by the method of (C), For example, what is shown with following general formulas (12), such as trimethoxysilylpropyl (meta) acrylate and methyl dimethoxy silyl propyl (meta) acrylate, is mentioned.
$H_2C=C(R^3)-R^4-R^{12}-[Si(R^1)_{2-b}(Y)_bO]-_m-Si(R^2)_{3-a}(Y)_a$ (12)
(R^1, R^2, R^3, R^4, Y, a, b, and m are the same as the above among a formula.) R^{12} may include one or more ether bonds by the divalent organic group of direct coupling or the carbon numbers 1-20. Especially although there is no restriction in particular at the stage to which the compound it has a compound, and the alkenyl group and crosslinkable silyl groups of polymerization nature in a monad is made to react, it is living radical polymerization, When it expects rubber character, it is preferred to make it react as the 2nd monomer aftar tha telophasa of a polymerization reaction or the ending reaction of a predetermined monomer. The hydrosilane etc. which have the mercaptan which has

crosslinkable silyl groups and crosslinkable silyl groups which are used by the chain transfer agent method of (D), and which are shown in JP,3-14068,B and JP,4-55444,B as a chain transfer agent which has crosslinkable silyl groups are mentioned.
[0037]The vinyl system polymer which is used by the method of (E) and which has at least one reactant high carbon-halogen bond can be obtained by the method of (E-a) - (E-b) as above-mentioned. What is shown by a general formula (13) as a compound having crosslinkable silyl groups and stabilization carbanion is mentioned into a monad.
$M^+C^-(R^7)(R^8)-R^{13}-C(H)(R^{14})-CH_2-[Si(R^1)_{2-b}(Y)_bO]_m-Si(R^2)_{3-a}(Y)_a$ (13)
(It is the same as R^1, R^2, R^7, R^8, Y, a, b, m, and ****** among a formula.) R^{14} in which R^{13} may include one or more ether bonds by the divalent organic group of direct coupling or the carbon numbers 1-10 shows hydrogen or the alkyl group of the carbon numbers 1-10, the aryl group of the carbon numbers 6-10, or the aralkyl group of the carbon numbers 7-10.
As an electron withdrawing group of R^7 and R^8, especially the thing that has the structure of $-CO_2R$, $-C(O)R$, and $-CN$ is preferred.
[0038]The vinyl system polymer which crosslinkable silyl groups has at the end of an at least one-piece chain, In order to obtain an organic halogenated compound or a sulfonyl halide compound by the polymerizing method using an initiator and a transition metal complex as a catalyst, It is preferred to use as an initiator the organic halogenated compound which has two or more reactant high carbon-halogen bonds used as the starting point, or a sulfonyl halide compound. As those examples, [0039]
[Formula 1]


[0040](R expresses the alkyl group of the carbon numbers 1-20, tha carbon number 6 - 20 aryl groups or the carbon number 7 - 20 aralkyl groups among a formula.) C_6H_4 expresses a phenylene

group. n expresses the integer of 0-20. X expresses chlorine, bromine, or iodine.
[0041]
[Formula 2]



[0042](X express chlorine, bromine, or iodine among a formula.) n expresses the integer of 0-20. C_6H_4 expresses a phenylene group, etc. — it is raised. Besides the above, the vinyl system polymer which has crosslinkable silyl groups at the end of an at least one-piece chain can be obtained. If the organic halogenated compound which has crosslinkable silyl groups is used as an initiator, the vinyl system polymer which has crosslinkable silyl groups in one end and in which other ends have the structure of the general formula 2 will be obtained. Thus, if halogen of the stop end of the polymer obtained is changed into a crosslinkable-silyl-groups content substituent, the vinyl system polymer which has crosslinkable silyl groups in both ends can be obtained. The already indicated method can be used as the converting method.
[0043]Although there is no restriction in particular as an organic halogenated compound which has crosslinkable silyl groups, what has the structure shown, for example in the general formula (14) and (15) is illustrated.
$R^{15}R^{16}C(X)-R^{17}-R^{18}-C(H)(R^{19})CH_2-[Si(R^1)_{2-b}(Y)_bO]_m-Si(R^2)_{3-a}(Y)_a$ (14)
(R^1, R^2, a, b, m, X, and Y are the same as the above among a formula.) Independently R^{15} and R^{16} Hydrogen or an alkyl group of the carbon numbers 1-20, An aryl group of the carbon numbers 6-20, an aralkyl group of the carbon numbers 7-20 or a thing mutually connected in the other end, and R^{17}

show -C(O) O-, -C(O)- or o-, m-, and p-phenylene group. R^{19} in which R^{18} may include one or more ether bonds by a divalent organic group of direct coupling or the carbon numbers 1-10 shows hydrogen or an alkyl group of the carbon numbers 1-10, an aryl group of the carbon numbers 6-10, or an aralkyl group of the carbon numbers 7-10.
$(R^2)_{3-a}(Y)_aSi-[OSi(R^1)_{2-b}(Y)_b]_m-CH_2-C(H)(R^{19})-R^{18}-C(R^{15})(X)-R^{17}-R^{16}$ (15)
(The inside of a formula, R^1, R^2, R^{15}, R^{16}, R^{17}, R^{18}, R^{19}, a, b, m, X, and Y are the same as the above) If an organic halogenated compound which has crosslinkable silyl groups is used as an initiator, a polymer whose one end is crosslinkable silyl groups and in which other ends are halogen will be obtained, but. A vinyl system polymer which has crosslinkable silyl groups in both ends can be obtained also by carrying out coupling of the halogen ends using a compound which can replace halogen of this polymer and which has a total of two or more same or different functional groups.
[0044]Although there is no restriction in particular as a compound which can replace end halogen and which has a total of two or more same or different functional groups, polyol, polyamine, polycarboxylic acid, polythiol and those salts, an alkaline metal sulfide, etc. are preferred. If an organic halogenated compound which has an alkenyl group is used for an initiator, it will have an alkenyl group in one end, and a polymer in which other ends are halogen will be obtained. Thus, if halogen of an end of a polymer obtained is changed into an alkenyl content substituent, If a vinyl system polymer which has an alkenyl group in both ends can be obtained and this alkenyl group is changed into crosslinkable silyl groups by an above-mentioned method etc., a vinyl system polymer which has crosslinkable silyl groups in both ends can be obtained.
[0045]In a use as which rubber character is required, since a molecular weight between the points constructing a bridge which has big influence on rubber elasticity can take greatly, as for at least one of crosslinkable silyl groups, it is preferred that it is in an end of a chain, and it is still more preferred that it is in molecular chain terminals altogether. Therefore, as for a vinyl system polymer which is used when compounding a vinyl system polymer which at least one crosslinkable silyl groups has and which has a hydroxyl group, halogen, or at least one alkenyl group, it is preferred that these functional groups are what exists in an end of a chain.
[0046]Although a vinyl system polymer which has at least one crosslinkable silyl groups, and a method of manufacturing an acrylic polymer especially (meta) are indicated by JP,3-14068,B, JP,4-55444,B, JP,6-211922,A, etc., for example, Since these methods use a "chain transfer agent method", such crosslinkable silyl groups exists in an end at a comparatively high rate, but there is a problem that molecular weight distribution of a polymer obtained is generally as large as two or more, and viscosity becomes high. Therefore, it is preferred to use a living-radical-polymerization method for obtaining a vinyl system polymer with low viscosity with narrow molecular weight distribution which has crosslinkable silyl groups at the end at a high rate. Even if it uses a condensation catalyst in stiffening an adhesive hardenability constituent of this invention, it is not necessary to carry out. As a condensation catalyst, titanate; dibutyltin dilaurate, such as tetrabuthyl titanate and tetrapropyl titanate, Dibutyl tin diacetyl acetonato, dibutyltin maleate, dibutyltin diacetate, Dibutyl tin dimethoxide, dibutyltin oxide, carboxylate, carboxylic acid, or a reactant of hydroxyl group-containing compound, Organic tin compounds, such as tin octylate and naphthenic acid tin; Aluminum tris acetylacetonato, Organoaluminium compounds, such as aluminumtrisethylacetoacetate and diisopropoxy aluminum ethylacetoacetate; Zirconium tetra acetylacetonato zirconium TETORAISOPUROPOKISAIDO, Organic zirconium compounds, such as zirconium tetra butoxide; Organic-lead-compounds [, such as lead octylate,];, A butylamine, octylamine, dibutyl amine, monoethanolamine, Diethanolamine, triethanolamine, diethylenetriamine, triethylenetetramine, oleylamine, octylamine, cyclohexylamine, benzylamine, diethylamino propylamine, xylylene diamine, triethylenediamine, guanidine, Diphenylguanidine, 2,4,6-tris(dimethyl aminomethyl) phenol, Morpholine, N-methylmorpholine, 1, 3-diazabicyclo. (5, 4, 6) An amine compound of undecene 7 grade. Or those carboxylate;. a reactant of lauryl amine and tin octylate or a reactant of an amine compound and an organic tin compound like a mixture, and mixture; — low-molecular-weight-polyamide resin; obtained from superfluous polyamine and polybasic acid — superfluous polyamine and a reactant; amino group of an epoxy compound. What is necessary is just to use one sort of silanol catalysts, or two sorts or more with publicly known a silane coupling agent which it has, for example, gamma-aminopropyl trimethoxysilane, N-(beta-aminoethyl) aminopropyl methyl dimethoxysilane, etc. if needed. As for the

amount used, it is preferred to use crosslinkable silyl groups in the range of zero to 10 weight section to a vinyl system polymer which it has at least one piece. When an alkoxy group is used as the hydrolytic basis Y, it is preferred to use a curing catalyst only with this polymer, since the cure rate is slow.

[0047]The polymer by this invention itself an adhesion promoter Ceramics other than glass and glass. Since it is possible to make it paste up to a wide range material by having an adhesive property to metal etc. or using various primers, it is not necessarily required, but it is preferred to use in order to acquire a stable adhesive property over a various substrate, parts, a base material, and adherend. As an adhesion promoter, phenol, cresol, a xylenol, resorcinol, alkylphenol and denaturation phenol (for example, cashew oil denaturation phenol.) tall oil denaturation phenol etc. — etc. — a phenol system compound and formalin. A resol type obtained by a reaction with aldehyde system compounds, such as paraformaldehyde, or novolac type phenol resin; sulfur; A bisphenol A type epoxy resin, Bisphenol F type epoxy resin, novolak type epoxy resin, a glycidyl ether type epoxy resin of a bisphenol A propylene oxide addition, Epoxy resins, such as a hydrogenation bisphenol A type epoxy resin; Alkyl titanate, such as tetrabuthyl titanate. Aromatic polyisocyanate;gamma-aminopropyl trimethoxysilane, such as tolylene diisocyanate and diphenylmethane diisocyanate, gamma-aminopropyl triethoxysilane, gamma-aminopropyl methyl dimethoxysilane, N-(beta-aminoethyl)-gamma-aminopropyl trimethoxysilane, N-(beta-aminoethyl)-gamma-aminopropyl triathoxysilane, A compound which carries out a crosslinkable-silyl-groups owner to an amino group into monads, such as N-(beta-aminoethyl)-gamma-aminopropyl methyl dimethoxysilane; Gamma-glycidoxypropyltrimetoxysilane, Compound;gamma-mercapto propyltrimethoxysilane which carries out a crosslinkable-silyl-groups owner to an epoxy group into monads, such as gamma-glycidoxy propyltriethoxysilane and gamma-glycidoxy propyl methyldimethoxysilane, Compound;gamma-isocyanate propyltrimethoxysilane which carries out a crosslinkable-silyl-groups owner to a sulfhydryl group into monads, such as gamma-mercaptopropyl triethoxysilane and gamma-mercaptpropylmethyl dimethoxysilane, gamma-isocyanate propyl triethoxysilane, A compound which carries out a crosslinkable-silyl-groups owner to an isocyanate group into monads, such as gamma-isocyanate propylmethyl dimethoxysilane; A compound which carries out a crosslinkable-silyl-groups owner to an amino group into the above monads, and a compound which carries out a crosslinkable-silyl-groups owner to an epoxy group into a monad. Or a reactant of a compound which carries out a crosslinkable-silyl-groups owner to an isocyanate group into a monad; gamma-(meta) acryloxyprophyltrimethoxysilane, gamma-(meta) acryloxyprophyltriethoxysilane, Reactant [of a compound which carries out a crosslinkable-silyl-groups owner to an acryloxy (meta) group, and a compound which carries out a crosslinkable-silyl-groups owner to an amino group into the above monads]; etc. are mentioned into monads, such as gamma-(meta)acryloxypropylmethyldimethoxysilane. These may be used alone or may be used together two or more kinds. A compound which carries out a crosslinkable-silyl-groups owner to an amino group especially into physical properties and a monad in which adhesive control is comparatively easy, A compound which carries out a crosslinkable-silyl-groups owner to an epoxy group into a monad, a compound which carries out a crosslinkable-silyl-groups owner to a sulfhydryl group into a monad, A reactant of a compound which carries out a crosslinkable-silyl-groups owner to an amino group into a monad, and a compound which carries out a crosslinkable-silyl-groups owner to an epoxy group into a monad, A compound which has an organic group which has at least one of nitrogen, oxygen, and sulfur atoms, and crosslinkable silyl groups in monads, such as a reactant etc. of a compound which carries out a crosslinkable-silyl-groups owner to an acryloxy (meta) group into a monad, and a compound which carries out a crosslinkable-silyl-groups owner to an amino group into a monad, is preferred. A compound in which the above-mentioned nitrogen, oxygen, and an organic group that has at least one of sulfur atoms have an organic group which is a basis generated when an amino group, an isocyanate group, or these react, and which has a nitrogen atom in a monad, and crosslinkable silyl groups from adhesive height is still more preferred.

[0048]As for the above-mentioned adhesion promoter, it is preferred that 20 weight sections are used from 0.01 to vinyl system polymer 100 weight section which has at least one crosslinkable silyl groups. In 0.01 weight sections, it is hard to reveal an adhesive improvement effect, and has an adverse effect on the physical properties of 20 weight-section ******** hardened material. An addition of an adhesion promoter is 0.5 to 5 weight section still more preferably 0.1 to 10 weight section preferably. In order to raise hardness when stiffening an adhesive hardenability constituent, or

to lower hardness, to take out elongation and to control physical properties, a physical-properties regulator can be used. As a physical-properties regulator, for example, methyl trimetoxysilane, dimethyldimethoxysilane, Alkyl alkoxysilane, such as trimethylmethoxysilane and n-propyltrimethoxysilane; Dimethyldi iso propenoxysilane, Alkyl iso propenoxysilane, such as methyl TORIISO propenoxysilane and gamma-glycidoxy propylmethyl JIISO propenoxysilane; Vinyltrimetoxysilane, Various silane coupling agents, such as vinylmethyldimethoxysilane, and silicone varnishes; polysiloxanes are added if needed. Vinyl system polymer 100 weight section [for] which has at least one crosslinkable silyl groups, if it adds in the range of zero to 20 weight section, a desirable result will be obtained. In order to speed up or delay a cure rate of an adhesive hardenability constituent and to suppress thickening under storage for a hardenability regulator again, a storage stability improving agent can be added. As a hardenability regulator or a storage stability improving agent, Alcohols; methyl orthoformate NADONO ortho ester, such as methanol and ethanol; carboxylic acid, such as compound;2-ethylhexanoic acid which carries out a crosslinkable-silyl-groups owner, such as a tetraethoxysilane, methyl trimetoxysilane, and vinyltrimetoxysilane, is mentioned. Vinyl system polymer 100 weight section [for] which has at least one crosslinkable silyl groups, if it adds in the range of zero to 20 weight section, a desirable result will be obtained.

[0049]In addition to this, an adhesive hardenability constituent of this invention Silica, carbon black, Various bulking agents, such as calcium carbonate; Aromatic dibasic acid esters, such as di(2-ethylhexyl) phthalate. Non-aromatic dibasic acid esters, such as dioctyl adipate; Polyether, such as a polypropylene glycol. Various plasticizers, such as acrylic oligomer; Various solvent; several-kinds silane coupling agents, such as toluene and methyl ethyl ketone, Various denaturing agents, such as a polysiloxane which has crosslinkable silyl groups; A polyamide wax, Colorant, such as the surface characteristic of rheology characteristic regulator; ultraviolet curing nature resin, such as hydrogenation castor oil and metallic soap, oxygen hardening resin, etc. and/or weatherproof improving agent; paints, and a color; additive agents, such as an antiaging agent, an ultraviolet ray absorbent, a SadamuMitsuyasu-ized agent, and a flameproofing agent, may also be used arbitrarily.

[0050]If a filler which can be added for the purpose of adjustment of a mechanical physical property is described in more detail when using an adhesive hardenability constituent of this invention as a sealing material composition, Fumed silica, sedimentation nature silica, a silicic acid anhydride, hydrous silicic acids, and a reinforcing filler like carbon black; Calcium carbonate, Fillers, such as magnesium carbonate, diatomite, calcination clay, clay, talc, titanium oxide, bentonite, organic bentonite, ferric oxide, a zinc oxide, an active white, and a milt balloon; asbestos, glass fiber, and a fibrous filler like a filament can be used. To obtain a hardened material with high intensity with these fillers. Mainly Fumed silica, sedimentation nature silica, a silicic acid anhydride, hydrous silicic acids, carbon black, A desirable result will be obtained if a filler chosen from surface treatment detailed calcium carbonate, calcination clay, clay, an active white, etc. is added in the range of one to 200 weight section to vinyl system polymer 100 weight section which has crosslinkable silyl groups. When elongation wants to obtain a hardened material which is size with low strength, A desirable result will be obtained if a filler mainly chosen from titanium oxide, calcium carbonate, talc, ferric oxide, a zinc oxide, a milt balloon, etc. is added in the range of one to 200 weight section to vinyl system polymer 100 weight section which has crosslinkable silyl groups. These fillers may be used by one kind and may carry out two or more kind mixing use.

[0051]If a plasticizer which can be added for adjustment of physical properties and viscosity is described in more detail, Dibutyl phthalate, diheptylphthalate, di(2-ethylhexyl) phthalate, Phthalic ester, such as di-isodecyl phthalate and butylbenzyl phthalate; Dioctyl adipate, Non-aromatic dibasic acid esters, such as dioctyl sebacate; Diethylene glycol dibenzoate, Ester species of polyalkylene glycols, such as TORIECHIRENGURIKORUJIBENZOETO; Tricresyl phosphate, Phosphoric ester, such as tributyl phosphate; Poly ERIREN glycol, polyether; which changed polypropylene glycols or these hydroxyl groups — chloroparaffin; — hydrocarbon system oils, such as alkyl diphenyl and partially-hydrogenated terphenyl, etc. are mentioned — these — independence — or, although two or more sorts can use it, mixing, It does not necessarily need. These plasticizers can also be blended at the time of polymer manufacture. A desirable result will be obtained if the amount of plasticizers is added in the range of zero to 100 weight section to vinyl system polymer 100 weight section which has crosslinkable silyl groups. A sealing material composition of this invention carries out combination seal preservation of all the combination ingredients beforehand, It is also possible to prepare as one

component type hardened by absorbing hygroscopic surface moisture in the after-construction air, ingredients, such as a curing catalyst, a filler, a plasticizer, and water, are separately blanded as a hardening agent, and it can also adjust as two component types mixed before using this compounding agent and a polymer composition. Hendling is easy and one component type also with few mistakes et the time of construction is more preferred.

[0052]Since it is what uses a vinyl system polymer as the main ingredients when using an adhesive hardenability constituent of this invention as a pressure sensitive adhesive oomposition, there is not necessarily necessity of adding a tackifier, but various kinds of things can be used if needed. If an example is given, they will be phenol resin, modified phenol resin, cyclopentadiene phenol resin, xylene resin, coumarone resin, petroleum resin, terpene resin, terpene phenol resin, rosin ester resin, etc.

[0053]If a solvent used in order to adjust workability is described in more detail, For example, ketones, such as ester solvents, such as aromatic hydrocarbons solvents, such as toluene and xylene, ethyl acetate, butyl acetate, emyl acetate, and cellosolve acetate, methyl ethyl ketone, methyl isobutyl ketone, and diisobutyl ketone, etc. ere mentioned. Those solvents may be used at the time of manufacture of a polymer.

[0054]Seid pressure sensitive adhesive composition is widely applicable to a tape, a sheet, a label, foil, etc. For example, a film made from a product made of a synthetic resin, or a conversion naturel product, paper, all kinds of cloth, applying said pressure sensitive adhesive composition to aubstrate materials, suoh as a metallic foil, metalizetion plastic foil, aabestos, or glass tissue, in a form of a solvent type, an emulsion type, or a hot melt type, and being exposed to humidity or moisture — room temperature setting -- or what is necessary is just to carry out heat cure When an adhesive hardenability constituent of this invention is used as a constituent for paints of a high solid, To a mathod desirable as a method of obtaining a vinyl system polymer which has at least one crosslinkable silyl groups. Since the manufacturing method is simple, a method of of (C) which carries out copolymerization of the compound having en alkenyl group and crosslinkable silyl groups of polymerization nature to other vinyl system monomers is also mentioned into a monad shown by a general formula (12). High solid-ization of a paint will be attained if these ere used.

[0055]Also in a compound it has a compound, and an alkenyl group and crosslinkable silyl groups of polymerization nature in a monad shown by a general formula (12). A compound especially whose crosslinkable silyl groups is alkoxy silyl groups is preferred in respect of cost or stability. For example, CH_2=$CHCO_2(CH_2)_3Si(OCH_3)_3$, CH_2=$CHCO_2(CH_2)_3Si(CH_3)(OCH_3)_2$, CH_2=$C(CH_3)CO_2(CH_2)_3Si(OCH_3)_3$ and especially CH_2=$C(CH_3)CO_2(CH_2)_3Si(CH_3)(OCH_3)_2$ are preferred. These compounds may be used independently, or two or more kinds may be mixed and they may be used.

[0056]although there is no restriction in perticular as a compound it has e compound, and an alkenyl group and crosslinkable silyl groups of polymerization nature in a monad, and a copolymerization ratio of other vinyl system monomers -- the oompound -- undar [a total polymerization presentation] and 1-50-mol % — desirable — 2-40-mol % — it is still more preferably [3-30 mol% of] good. If hardening becomes thet quantity of a compound it has a compound, and an alkanyl group and crosslinkable silyl groups of polymerization nature in a moned is less than [1 mol %] insuffioient and 50-mol % is exceeded, storage stability will worsen.

[0057]An above-mentioned polymerization method is the polymerization controlled as already stated, and can obtain a narrow vinyl system polymer of molecular weight distribution. Since molecular weight distribution is narrow, viscosity of a polymer is stopped low, and it becomes possible to give plasticity required of a paint with a smeller solvent amount. In addition to an additive egent described as an adhesive hardenability constituent, to this constituent for paints, additive agents, such as resin, such as polyester, epoxy, and an acrylic, a coloring assistant, a flowing agent, a defoaming agent, a spray for preventing static electricity, can edd ** if needed. If colorant used for this constituent for paints is described in more detail, organic colors, such as inorganic pigments, such as a titanium dioxide, carbon black, iron oxide, and chrome oxide, a phthalocyanine system, and a quinacridone series, etc. will be mentioned. A blending ratio of these additivea can be suitably chosen according to the required characteristic, and mixing and using is also possible. A uniform coat can be obtained, if a vinyl aystem polymer which has at least one crosslinkeble silyl groupa shown in a general formula (1) is stiffened after adding if needed end painting a curing catelyst and an additive agent to a coated object. Since it goes on at a room temperature, it is not neceasary to haat in the case of hardening but, and hydrolysis and/of crosslinkeble ailyl groupa, or condensation may be heated for promotion of

hardening. 20-200 ** of cooking temperature is 50-180 ** preferably.

[0058]A constituent for painta of this invention can be used as a solvent system or a drainage system paint. It is also possible to distill volatile matter content out of a vinyl system polymar which ia the main ingredients, to pulverize a compound, after adding a desired compound, and to uae it as powder coatings. When an adhesive hardenability oonstituent of this invention is used as a constituent for paints excellent in character possibla [high-aolid-izing] and alastic, Although it is preferred that it is in molecular chain terminals as for at least one of crosslinkable silyl groups, In order to adjust a molecular weight between the points constructing e bridge, crosslinkable silyl groups may be introduced into ** in a chain by carrying out copolymerization of a little compounds it hes compounds, and an alkenyl group and crosslinkable silyl groups of polymerization nature in a moned shown by a generel formula (12). CH_2=$CHCO_2(CH_2)_3Si(OCH_3)_3$, CH_2=$CHCO_2(CH_2)_3Si(CH_3)(OCH_3)_2$, CH_2=$C(CH_3)CO_2(CH_2)_3Si(OCH_3)_3$, CH_2=$C(CH_3)CO_2(CH_2)_3Si(CH_3)(OCH_3)$ 2, CH2=$CHCO_2(CH_2)_3Si(OC_2H_5)_3$, CH_2=$CHCO_2(CH_2)_3Si(CH_3)(OC_2H_5)_2$, CH_2=$C(CH_3)CO_2(CH_2)_3Si(OC_2H_5)_3$, CH_2=$C(CH_3)CO_2(CH_2)_3Si(CH_3)(OC_2H_5)_2$, CH_2=$CHCO_2(CH_2)_3Si(OC_2H_5)_3$, CH_2=$CHCO_2(CH_2)_3Si(CH_3)(OC_2H_5)_2$, CH_2=$C(CH_3)CO_2(CH_2)_3Si(OC_2H_5)_3$, CH_2=$C(CH_3)CO_2(CH_2)_3Si(CH_3)(OC_2H_5)_2$ etc. can be illustrated. These compounds may be used independently, or two or more kinds may be mixed and they may be used. In addition to an additive agent deacribed as en adhesive hardanability constituent, to this conatituent for paints, edditive agants, such as resin, such as polyester, epoxy, and an aorylio, a coloring assistant, a flowing agent, a defoaming agent, a aprey for preventing static electricity, can add ** if needad. If colorant usad for this constituent for paints is described in more detail, organic colors, such as inorganio pigments, such as a titanium dioxida, carbon black, iron oxide, and chrome oxide, a phthelocyanine system, end a quinacridone series, etc. will be mentioned. A blending ratio of these additives can be suitably chosen according to the required characteristic, and mixing and using is also possible. A uniform coat can be obtained, if a vinyl system polymer which has at least one crosslinkable silyl groups shown in a general formula (1) is stiffened after adding if needed and painting a curing catalyst and an additive agent to a coated object. Since it goes on at a room temperature, it is not necessary to heat in the case of hardening but, and hydrolysis and/of crosslinkable silyl groups, or condensation mey be heated for promotion of hardening. 20-200 ** of cooking temperature is 50-180 ** preferably. A constituent for paints of this invention can be used as a solvent system or a drainage system paint. It is also possible to distill volatile matter content out of a vinyl system polymer which is the main ingredients, to pulverize a compound, after adding a desired compound, and to use it as powder coatings.

[0059]Although this invention is explained based on an example below, it ia not limited to the following example.

(Raference example 1) The bottom of a synthetio nitrogen atmosphera of a hydroxyl group content initiator, ethylene glyool (10.9 mL) 2-bromo propionyl chlorida (2mL, 3.35 g, 19.5mmol) was slowly dropped at a THF aolution (10mL) of 195mmol and pyridine (3 g, 39mmol) at 0 **. A aolution was agitated et a tampereture as it is for 2 hours. Dilute hydrochloric ecid and athyl acetate were added, two-layer was separated, and an organic layer was dried by washing and Na_2SO_4 by dilute hydrochloric acid and brine. Volatile matter content was distilled off under decompression and partially purified substance wes obtained (3.07g). Hydroxyethyl 2-bromo propionate shown in (70-73 **, 0.5mmHg), and a lower type was obtained by carrying out distillation under reduced pressure of this partially purified substance (2.14 g, 56%).

$H_3CC(H)(Br)C(O)O(CH_2)_2$-OH (synthetic example 1)

To a resisting pressure reaction vessel of the poly (acrylic acid-n-butyl) composition 1L which has e hydroxyl group at the end. Acrylic ecid-n-butyl (112mL, 100 g, 0.78 mol), A hydroxyl group content initiator obtained by the reference example 1 (3.07 g, 15.6mmol). Copper (2.24 g, 15.6mmol), a 2,2'-bipyridyl (4.87 g, 31.2mmol), ethyl ecetate (90mL), and acetonitrile (20mL) were taught for a start [bromination], and the sealed tube was carried out, after performing nitrogen bubbling end removing dissolved oxygen. A mixture was heated at 130 ** and made to react for 2 hours. A reaction veasel was returned to a room temperature, methacrylic acid-2-hydroxyethyl (3.92mL, 4.06 g, 31.2mmol) was added, and it waa meda to react at 110 ** for 2 houra. After diluting a mixture with ethyl acetate (200mL) and filtering insoluble metter, it waahed by chloride and brine 10%, and filtrate was dried for

an organic layer by Na_2SO_4. A solvent was distilled off under decompression and 82g of poly (acrylic acid-n-butyl) which has a hydroxyl group at the end was obtained. Viscosity of this polymer was 25 Pa-s, by GPC measurement (mobile phase chloroform, polystyrene conversion), a number average molecular weight was 5100 and molecular weight distribution was 1.29. The number of a hydroxyl group of an average per one molecule of polymers for which it asked from ^{1}H-NMR analysis was 2.39 pieces.
Azeotropy drying was performed for poly (acrylic acid-n-butyl) (4.94 g, OH=2.30mmol) which has a hydroxyl group at the end compounded by the poly (acrylic acid-n-butyl) synthetic above which has crosslinkable silyl groups at the end at 50 ** under toluene existence. Octylic acid tin (4.9 mg) and toluene (6mL) were added here, and a methyl dimethoxy silyl propylisocyanate (0.524 g, 2.77mmol) was dropped at 50 **. After an end of dropping, reaction temperature was raised to 70 ** and a reaction was continued for 4 hours. When a signal (3.8 ppm) of a methylene group which a hydroxyl group combined by ^{1}H-NMR disappeared, it was judged as what an unreacted hydroxyl group does not have. Volatile matter content was distilled off with decompression and poly (acrylic acid-n-butyl) which has crosslinkable silyl groups at the end was obtained. Viscosity of this polymer was 22 Pa-s, by GPC measurement (mobile phase chloroform, polystyrene conversion), a number average molecular weight was 4900 and molecular weight distribution was 1.60.
(Example 1) To polymer 100 weight section which has crosslinkable silyl groups at the end obtained in the synthetic example 1, dibutyltin diacetyl acetonato 1 weight section was mixed, and it slushed into a mold, and defoamed at a room temperature using a vacuum drier. A uniform rubber-like hardened material sheet was obtained by carrying out heat cure at 50 ** for 20 hours. A gel fraction for which it asked by toluene extraction was 93%. piercing 2 (1/3) item type dumbbell specimen from a rubber-like hardened material sheet, and pulling using an autograph — an examination — having carried out (200 mm/min) -- time — breaking strength -- 0.31MPa and elongation after fracture -- 35% -- it was .
(Synthetic example 2)
To a toluene solution (100mL) of poly (acrylic acid-n-butyl) (50g) and pyridine (10mL) which have a hydroxyl group at the end obtained in the poly (acrylic acid-n-butyl) example 1 of synthetic composition which has an alkenyl group at the end. An undecene acid chloride (7.22mL, 6.81 g, 33.6mmol) was slowly dropped at 75 ** under a nitrogen atmosphere, and it agitated at 75 ** for 3 hours. A generated white solid was filtered, an organic layer was washed by dilute hydrochloric acid and brine, and an organic layer was dried by Na_2SO_4. By condensing under decompression, poly (acrylic acid-n-butyl) (43g) which has an alkenyl group was obtained. By GPC measurement (mobile phase chloroform, polystyrene conversion), a number average molecular weight of a polymer was 5400, and molecular weight distribution was 1.30. The number of alkenyl groups introduced per [for which it asked from ^{1}H-NMR analysis] one molecule of polymers was 2.28.
To a resisting pressure reaction vessel of poly (acrylic acid-n-butyl) synthetic 30mL which has crosslinkable silyl groups at the end. Poly (butyl acrylate) (2g) which has an alkenyl group in both ends obtained above, Methyl dimethoxysilane (0.32mL), methyl orthoformate (0.09 mL) 3 Eq and a platinum screw (divinyl tetramethyl disiloxane) (it is the 10^{-4} equivalent to a $8.3x10^{-8}$mol/L xylene solution and an alkenyl group) were prepared to an alkenyl group, and it agitated at 100 ** for 1 hour. By distilling off volatile matter content under decompression, 2g of poly (acrylic acid-n-butyl) which has crosslinkable silyl groups was obtained. By GPC measurement (mobile phase chloroform, polystyrene conversion), a number average molecular weight of a polymer was 5900, and molecular weight distribution was 1.37. The number of silyl groups introduced per [for which it asked from ^{1}H-NMR analysis] one molecule of polymers was 2.24.
(Example 2) A crosslinkable-silyl-groups end polymer (1g) and a curing catalyst (Japanese east transformation make, U-220, dibutyltin diacetyl acetonato, 30 mg) of the synthetic example 2 were mixed well, and it slushed into a mold, and defoamed at a room temperature using a vacuum drier. A uniform rubber-like hardened material was obtained by neglecting it for seven days to a room temperature. A gel fraction for which it asked by toluene extraction was 78%.
(Example 3) Polymer 100 weight section which has crosslinkable silyl groups at the end of the synthetic example 2, water 1 weight section, and dibutyltin JIMETOKI side 1 weight section were

mixed well, and it slushed into a mold, and defoamed at a room temperature using decompression oven. A uniform rubber-like hardened material sheet was obtained by carrying out heat cure at 50 ** for 20 hours. A gel fraction for which it asked by toluene extraction was 88%. 2 (1/3) item type dumbbell specimen was pierced from a rubber-like hardened material sheet, and it examined by pulling using an autograph (200 mm/min). Breaking strength was 0.32MPa and elongation after fracture was 34%.
(Synthetic example 3)
To a resisting pressure reaction vessel of 500 ml of poly (acrylic acid-n-butyl) composition which has halogen at the end. Acrylic acid-n-butyl (112mL, 100 g, 0.78 mol), Dibromoxylene (4.12 g, 15.6mmol), the first copper of bromination (2.24 g) 15.6mmol, a 2,2'-bipyridyl (4.87 g, 31.2mmol), ethyl acetate (90mL), and acetonitrile (20mL) were taught, and the sealed tube was carried out, after performing nitrogen bubbling and removing dissolved oxygen. A mixture was heated at 130 ** and made to react for 2 hours. A reaction vessel was returned to a room temperature, methacrylic acid-2-hydroxyethyl (3.92mL, 4.06 g, 31.2mmol) was added, and it was made to react at 110 ** for 2 hours. A mixture was diluted with ethyl acetate (200mL), and a polymer which has Br basis at the end was obtained by carrying out removal refining of the copper catalyst through a column of activated alumina. Number average molecular weights of an obtained polymer were 5700 and molecular weight distribution 1.37 in GPC measurement (mobile phase chloroform, polystyrene conversion). Under a poly (acrylic acid-n-butyl) synthetic nitrogen atmosphere which has an alkenyl group at the end,84 g of poly (acrylic acid-n-butyl) which has halogen, the pentene acid potassium 7.7g (56mmol), and DMAc80ml were taught to an end obtained above, and it was made to react to a 500-ml flask at 70 ** for 4 hours. Water extraction refining removed unreacted pentene acid potassium and generated potassium bromide in reaction mixed liquor, and a polymer which has an alkenyl group at the end was obtained. Silicic acid aluminum (a product made from harmony chemicals: ****- word 700PEL) of this polymer 70g, this, and same weight was mixed in toluene, and it agitated at 100 **. Silicic acid aluminum was filtered 4 hours afterward, and a polymer was refined by heating volatile matter content of filtrate under decompression, and distilling it off. Number average molecular weights of an obtained polymer were 4760 and molecular weight distribution 1.73 in GPC measurement (mobile phase chloroform, polystyrene conversion). The number of an alkenyl group per one molecule of polymers for which it asked from ^{1}H-NMR analysis was 1.78 pieces.
The polymer 60g which has an alkenyl group at the end obtained above to poly (acrylic acid-n-butyl) 200 ml of composition resisting pressure coil which has crosslinkable silyl groups at the end, Taught methyl dimethoxysilane 8.4mL (68.1mmol), methyl orthoformate 2.5mL (22.9mmol), and platinum bis (divinyl tetramethyl disiloxane)$5x10^{-3}$mmol, it was made to react at 100 ** for 4 hours, and a cross-linking silicon group content polymer was obtained. Number average molecular weights of an obtained polymer were 6000 and molecular weight distribution 1.44 in GPC measurement (mobile phase chloroform, polystyrene conversion). The number of crosslinkable silyl groups per one molecule of polymers for which it asked from ^{1}H-NMR analysis was 1.59 pieces.
(Example 4) Mixed stirring of water 1 weight section and the dibutyl tin JIMETOKI side 1 weight section was carried out at polymer 100 weight section which has crosslinkable silyl groups at the end obtained in the synthetic example 3, and it slushed into a 2-mm-thick mold. A uniform rubber-like hardened material sheet was obtained by defoaming at a room temperature using a vacuum drier, and carrying out heat cure for two days at 50 **. A gel fraction for which it asked by toluene extraction was 93%. 2 (1/3) item type dumbbell specimen was pierced from a rubber-like hardened material sheet, and it examined by pulling using an autograph (200 mm/min). Breaking strength was 0.26MPa and elongation after fracture was 75%.
(Synthetic example 4)
In poly (acrylic acid-n-butyl) 50 ml of composition flask which has halogen at the end, the first copper 0.63g (4.4mmol) of bromination, 0.76 g (4.4mmol) of pentamethyl diethylenetriamine, After teaching 5 ml of acetonitrile, 1.6 g (4.4mmol) of 2,5-dibromo diethyl adipate, and 44.7 g (349mmol) of butyl acrylate and performing freezing deaeration, 70 ** was made to react under a nitrogen atmosphere for 7 hours. A polymer which has Br basis at the end was obtained by carrying out removal refining of the copper catalyst through a column of activated alumina. Number average molecular weights of an obtained polymer were 10700 and molecular weight distribution 1.15 in GPC measurement (mobile phase chloroform, polystyrene conversion).

Under a poly (acrylic acid-n-butyl) synthetic nitrogen atmosphere which has an alkenyl group at the end, 35 g of poly (acrylic acid-n-butyl) which has halogen, the pentene acid potassium 2.2g (16.1mmol), and DMAc35mL were taught to an end obtained above, and it was made to react to a 200-ml flask at 70 ** for 4 hours. Water extraction refining removed unreacted pantene acid potassium and generated potassium bromide in reaction mixed liquor, and a polymer which has an alkenyl group at the end was obtainad. Number average molecular weights of an obtainad polymer were 11300 and molecular weight distribution 1.12 in GPC measurement (mobile phase chloroform, polystyrene conversion). The number of an alkenyl group per one molecule of polymers for which it asked from ^{1}H-NMR analysis was 1.82 pieces.
The polymer 15g which has an alkenyl group at the end obtained above to a poly (acrylic acid-n-butyl) synthetic 200mL resisting pressure coil which has crosslinkable silyl groups at the end, Taught methyl dimethoxysilane 1.8mL (14.5mmol) methyl orthoformate 0.26mL (2.4mmol) and platinum bis (divinyl tetramethyl disiloxane)10^{-4}mmol, it was made to react at 100 ** for 4 hours, and a polymer which has crosslinkable silyl groups at the end was obtained. Viscosity of an obtained polymer was 44 Pa-s, and number average molecular weights were 11900 and molecular weight distribution 1.12 in GPC measurement (mobile phase chloroform, polystyrene conversion). The number of a cross-linking silicon group per one molecule of polymars was 1.46 pieces by ^{1}H-NMR analysis.
(Example 5) Mixed stirring of watar 1 weight saction and the dibutyltin JIMETOKI side 1 weight section was carried out at polymer 100 weight section which has crosslinkabla silyl groupa at the end obtained in the synthetic axample 4, and it slushed into a 2-mm-thick mold. A uniform rubber-like hardened material sheat was obtainad by defoaming at a room temperature using decompression oven, and carrying out heat oure for ten days at 50 **. A gel fraction for which it asked by torr ENN extraction was 98%. 2 (1/3) item typa dumbbell specimen was pierced from a rubber-like hardened material sheet, and it examined by pulling using an autograph (200 mm/min). Breaking strength was 0.35MPa and elongation after fracture was 77%.
(Synthetic example 5)
To a glass reaction vessel of poly (acrylic acid-n-butyl) synthetic 100mL which has an alkenyl group at the end. Butyl acrylate (50.0mL, 44.7 g, 0.349 mol), Copper (1.25 g, 8.72mmol), pentamethyl diethylenetriamine (1.82mL, 1.51 g, 8.72mmol), and acetonitrile (5mL) were taught for a start [bromination], and nitrogen gas replaced, after carrying out after-cooling decompression deaeration. After agitating well, a diethyl 2,5-dibromo horse mackerel peat (1.57 g, 4.36mmol) was added, and heating churning was carried out at 70 **. 1 and 7-octadien (6.44mL, 4.80 g, 43.6mmol) was added 60 minutes afterward, and heating churning was continued at 70 ** for 2 hours. After processing a mixture with activated alumina, volatile matter content was heated under decompression and distilled off. Output was dissolved in ethyl acetate and it washed by chloride and brine 2%. A polymer which has an alkenyl group at the end was obtainad by drying an organic layer by Na_2SO_4, heating volatile matter content under decompression and distilling it off. By GPC measurement (polystyrene conversion), a number average molecular weight of an obtained polymer was 13100, and molecular weight distribution was 1.22. The number of an alkenyl group per one molecule of polymers for which it asked from ^{1}H-NMR analysis was 2.01 pieces.
. Were obtained by the poly (acrylic acid-n-butyl) synthetic above which has crosslinkable silyl groups at the end. Silicic acid aluminum (a product made from harmony chemicals: ****- word 700PEL) of same weight was mixed with poly (acrylic acid-n-butyl) (30.5g) and a polymer which have an alkenyl group at the end in toluene, and it agitated at 100 **. Silicic acid aluminum was filtered 4 hours afterward, and a polymer was refined by heating volatile matter content of filtrate under decompression, and distilling it off. The refined above-mentioned polymer (23.3g), dimethoxymethylsilane (2.55mL, 20.7mmol), alt.formic acid dimethyl (0.38mL, 3.45mmol), and a platinum catalyst were taught to a resisting pressure glass reaction vessel of 200mL. However, the amount of platinum catalyst used was made into the $2x10^{-4}$ equivalent by a mole ratio to an alkenyl group of a polymer. A reaction mixtura was heated at 100 ** for 3 hours. By carrying out decompression distilling off of the volatile matter content of a mixture, poly (acrylic acid-n-butyl) which has crosslinkable silyl groups at the end was obtained. Number average molecular weights of an obtained polymer ware 13900 and molecular weight distribution 1.25 in GPC measurement (mobile phase ohloroform, polystyrena conversion). The number of a cross-linking silicon group per one

molecule of polymers for which it asked from ^{1}H-NMR analysis was 1.58 pieces.
(Example 6) Mixed stirring of water 1 weight section and the dibutyl tin JIMETOKI side 1 weight section was carried out at polymer 100 weight section which has crosslinkable silyl groups at tha end obtained in the synthetic example 5, and it slushed into a 2-mm-thick mold. A uniform rubber-like hardened material sheet was obtained by defoaming at a room temperature using a vacuum drier, and carrying out heat cure for ten days at 50 **. A gel fraction for which it asked by toluene extraction was 85%. 2 (1/3) item typa dumbbell specimen was pierced from a rubber-like hardened material sheet, and it examined by pulling using an autograph (200 mm/min). Breaking strength was 0.34MPa and elongation after fracture was 86%.
(Synthetic example 6)
In poly (acrylic acid-n-butyl) synthetic 50mL flask which has halogen at the end, the first copper 0.63g (4.4mmol) of bromination, 0.76 g (4.4mmol) of pentamethyl diethylenetriamine, After teaching acetonitrile 5mL, 0.78 g (2.2mmol) of 2,5-dibromo diethyl adipate, and 44.7 g (349mmol) of butyl acrylate and performing freezing deaeration, 70 ** was made to react under a nitrogen atmosphere for 6 hours. A polymer which has Br basis at the end was obtained by carrying out removal refining of the copper catalyst through a column of activated alumina. Number average molecular weights of an obtained polymer were 23600 and molecular weight distribution 1.14 in GPC measurement (mobile phase chloroform, polystyrene conversion).
The polymer 34g which haa Br basis, the pentene acid potassium 1.0g (7.6mmol), and DMAc34mL were taught to an end obtained above in a 200mL flask under a poly (acrylic acid-n-butyl) synthetic nitrogen atmosphere which has an alkenyl group at the end, and it was made to raact at 70 ** for 4 hours. Water extraction refining removed unreacted pentene acid potassium and generated potassium bromide in reaction mixed liquor, and a polymer which has an alkanyl group at the end was obtained.
A polymer and silicic acid aluminum (a product made from harmony chemicals: ****- word 700PEL) of same weight (30.5g) which have an alkenyl group at this end were mixed in toluene, and it agitated at 100 **. Silicic acid aluminum was filtered 4 hours afterward, and a polymer was refined by heating volatile matter content of filtrate under decompression, and distilling it off. Number average molecular weights of an obtained polymer were 24800 and molecular weight distribution 1.14 in GPC measurement (mobile phase chloroform, polystyrene conversion). The number of an alkenyl group per one molecule of polymers for which it asked from ^{1}H-NMR analysis was 1.46 pieces.
The polymer 21g which has an alkenyl group at the end obtained above to poly (acrylic acid-n-butyl) 200 ml of composition resisting pressure coil which has crosslinkable silyl groups at the end, Taught 0.13 ml (1.3mmol) of 0.94 ml (7.6mmol) of methyl dimethoxysilane methyl orthoformate, and platinum bis(divinyl tetramethyl disiloxane)$2x10^{-4}$mmol, it was made to react at 100 ** for 4 hours, and a polymer which haa crosslinkable silyl groups at the end was obtained. Viscosity of an obtained polymer was 100 Pa-s, and number average molecular weights were 25400 and molecular weight distribution 1.16 in GPC measurement (mobile phasa chloroform, polystyrene conversion). The number of crosslinkable silyl groups per one molecule of polymers for which it asked from ^{1}H-NMR analysis was 1.48 pieces.
(Example 7) Mixed stirring of water 1 weight section and the dibutyl tin JIMETOKI side 1 weight section was carried out at polymer 100 weight section which has crosslinkable silyl groups at the end obtained in the synthetic example 6, and it slushed into a 2-mm-thick mold. A uniform rubber-like hardened material sheet was obtained by defoaming at a room temperature using decompression oven, and carrying out heat cure for two days at 50 **. A gel fraction for which it asked by toluene extraction was 94%. 2 (1/3) item type dumbbell specimen was pierced from a rubber-like hardened material sheet, and it examined by pulling using an autograph (200 mm/min). Breaking strength was 0.40MPa and elongation after fracture was 323%.
(Example 1 of comparison composition)
According to Example 1 of poly (acrylic acid-n-butyl) synthetic JP,5-262808,A which has a hydroxyl group at the end using hydroxyl group content disulfide, 2-hydronalium KISHIE ethyldisulfide (30.8 g, 0.2 mol) was added to a flask of 100mL. A flask was heated at 100 ** and a mixture of acrylic acid-n-butyl (12.8 g, 0.1 mol) and azobisisobutyronitrile (0.328 g, 0.002 mol) was dropped over 30 minutes. A mixture was agitated at 100 ** for further 1 hour. Toluene (20mL) was added, a mixture was settled on a separating funnel, and a lower layer was separated. After washing the upper layer 3 times

with water and drying by Na_2SO_4, poly (acrylic acid-n-butyl) which has a hydroxyl group in both ends was obtained by distilling off volatile matter content under decompression (12.2 g, 95%). Viscosity of this polymer was 49 Pa-s, by GPC measurement (mobile phase chloroform, polystyrene conversion), a number average molecular weight was 4200 and molecular weight distribution was 4.16. The number of a hydroxyl group of an average per one molecule of polymers for which it asked from ^{1}H-NMR analysis was 1.42 pieces.
Azeotropy drying was performed for poly (acrylic acid-n-butyl) (4.52 g, OH=1.85mmol) which has a hydroxyl group at the end compounded by the poly (acrylic acid-n-butyl) synthetic above which has crosslinkable silyl groups at the end using hydroxyl group content disulfide at 50 ** under toluene existence. Octylic acid tin (4.52 mg) and toluene (6mL) were added here, and a methyl dimethoxy silyl propylisocyanate (0.421 g, 2.22mmol) was dropped at 50 **. After an end of dropping, reaction temperature was raised to 70 ** and a reaction was continued for 4 hours. When a signal (3.8 ppm) of a methylene group which a hydroxyl group combined by ^{1}H-NMR disappeared, it was judged as what an unreacted hydroxyl group does not have. Volatile matter content was distilled off with decompression and poly (acrylic acid-n-butyl) which has crosslinkable silyl groups at the end was obtained. Viscosity of this polymer was 53 Pa-s, by GPC measurement (mobile phase chloroform, polystyrene conversion), a number average molecular weight was 4700 and molecular weight distribution was 3.71.

(Comparative example 1) To polymer 100 weight section which has crosslinkable silyl groups at the end of the example 1 of comparison composition, dibutyltin diacetyl acetonato 1 weight section was mixed, and it slushed into a mold, and defoamed at a room temperature using decompression oven. A uniform rubber-like hardened material sheet was obtained by carrying out heat cure at 50 ** for 20 hours. A gel fraction for which it asked by toluene extraction was 82%. Although an extracted part was condensed and ^{1}H-NMR was measured, crosslinkable silyl groups did not exist in it.
[0060]piercing 2 (1/3) item type dumbbell specimen from a rubber-like hardened material sheet, and pulling using an autograph -- an examination — having carried out (200 mm/min) — time — breaking strength — 0.21MPa and elongation after fracture — 93% — it was .
(Example 2 of comparison composition)
400 g of poly (acrylic acid-n-butyl) synthetic toluene which has the crosslinkable silyl groups using a cross-linking silicon group content monomer. It polymerized at 105 ** for 7 hours, carrying out nitrogen bubbling of 385 g of butyl acrylate, 15 g of methyl methacrylate dimethoxy silyl propyl, and 6 g of the azobisisobutyronitrile in 1L flask. Poly (acrylic acid-n-butyl) which has crosslinkable silyl groups was obtained by distilling off toluene. Viscosity of this polymer was 74 Pa-s, by GPC measurement (mobile phase chloroform, polystyrene conversion), a number average molecular weight was 8500 and molecular weight distribution was 2.47. The number of a hydroxyl group of an average per one molecule of polymers for which it asked from ^{1}H-NMR analysis was 1.40 pieces.
(Comparative example 2) Mixed stirring of water 1 weight section and the dibutyl tin JIMETOKI side 1 weight section was carried out at polymer 100 weight section which has the crosslinkable silyl groups of the example 2 of comparison composition, and it slushed into a 2-mm-thick mold. A uniform rubber-like hardened material sheet was obtained by defoaming at a room temperature using decompression oven, and carrying out heat cure for ten days at 50 **. A gel fraction for which it asked by toluene extraction was 78%. 2 (1/3) item type dumbbell specimen was pierced from a rubber-like hardened material sheet, and it examined by pulling using an autograph (200 mm/min). Breaking strength was 0.14MPa and elongation after fracture was 69%.
(Comparative example 3)
210 g of poly (acrylic acid-n-butyl) synthetic toluene which has the crosslinkable silyl groups using a cross-linking silicon group content monomer. It polymerized at 105 ** for 7 hours, carrying out nitrogen bubbling of 293 g of butyl acrylate, 7.2 g of methyl methacrylate dimethoxy silyl propyl, and 1.8 g of the azobisiso valeronitrile in 1L flask. Poly (acrylic acid-n-butyl) which has crosslinkable silyl groups was obtained by distilling off toluene. Viscosity of this polymer was 110Paands, by GPC measurement (mobile phase chloroform, polystyrane conversion), a number average molecular weight was 9600 and molecular weight distribution was 2.86. A result of Examples 1-7 except Example 2 and the comparative examples 1-3 was summarized in Table 1.
[0061]

[Table 1]

	実施例1	実施例3	実施例4	実施例5	実施例6	実施例7	比較例1	比較例2	比較例3
重合体	合成例1	合成例2	合成例3	合成例4	合成例5	合成例6	比較合成例1	比較合成例2	比較例3
粘度(Pa·s)	22	–	–	44	–	100	53	74	110
Mn	4900	5900	6000	11900	13900	25400	4700	8500	9600
Mw/Mn	1.60	1.37	1.44	1.12	1.25	1.16	3.71	2.47	2.86
Fn	(2.39)	2.24	1.59	1.40	1.58	1.48	(1.42)	1.40	–
ゲル分率(%)	93	88	83	98	85	94	82	78	–
破断時強度(MPa)	0.31	0.32	0.20	0.35	0.34	0.40	0.21	0.14	–
破断時伸び(%)	35	34	75	77	86	323	93	69	–

Fn:一分子当たりの平均の架橋性シリル基の数
ただし、合成例1および比較例1の値は合成中間体である水酸基末端重合体中の水酸基の数を示す。

[0062]When it is a use as which rubber character is required, in order to raise balance of elongation at the time of intensity/fracture at the time of a modulus/fracture, polymers quantification of a polymer molecular weight is desired. In a polymer which has a cross-linking functional group like especially this invention, since a molecular weight is closely connected with a molecular weight between the points constructing a bridge used as an important index of a rubber design, the tendency is strong. A number average molecular weight is one of the important parameters which influence a molecular weight between the points constructing a bridge, and it is desirable for Kougami of the above-mentioned properties balance that this can be enlarged.
[0063]Since the vinyl system polymer which has the crosslinkable silyl groups of this invention can control molecular weight distribution narrowly, viscosity when the polymer of the almost same number average molecular weight compares becomes very low, and serves as a raw material which was excellent in handling nature as a result (viscosity of Example 1 below half of the comparative example 1). Example 5 and the comparative example 3 — the same . When viscosity has restriction, since a polymer with a higher number average molecular weight is compoundable, with the almost same viscosity, the hardened material which was more excellent in a modulus / intensity / elongation balance is obtained (example 7). Since the vinyl system polymer which has crosslinkable silyl groups by a living-radical-polymerization method has been obtained, Even if the quantity of the crosslinkable silyl groups of the average per monad is almost the same, the quantity of the polymer which does not contain crosslinkable silyl groups decreases, and the high hardened material for gel can be obtained (Example 5 and the comparative example 1).
(Example 8) Some poly (acrylic acid-n-butyl) hardened material sheets which have the crosslinkable silyl groups obtained in heat-resistant example 5 of the hardened material were put into 150 ** oven, it took out 24 hours afterward, and the surface state was observed. It was normal to a surface state.
(Example 3 of comparison composition)
In 200 ml of composition flask of the poly dimethylsiloxane (silicone) which has crosslinkable silyl groups at the end, 97 g of end vinylpoly dimethylsiloxane (DMS-V25 made from AZUMAKKUSU: unsaturation group equivalent 0.11 eq/kg) of the molecular weight 17,200, 2.3 g (21.4mmol) of methyl dimethoxysilane and platinum bis(divinyl tetramethyl disiloxane)10^{-3}mmol was added, and it reacted at 70 ** for 6 hours. The number average molecular weights of the obtained crosslinkable-silyl-groups end poly dimethylsiloxane were 11900 and molecular weight distribution 2.52 in GPC measurement (mobile phase chloroform, polystyrene conversion). The number of the cross-linking silicon group per one molecule of poly dimethylsiloxane polymer for which it asked from the intensity ratio of the methyl proton which the peak of unsaturation group origin disappeared by ^{1}HNMR (300 MHz), and was combined with the silicon atom of polymer backbone origin, and the proton of a methoxy silyl group was 2. Viscosity was 6 poise.
(Comparative example 4) Mixed stirring of water 1 weight section and the dibutyltin JIMETOKI side 1 weight section was carried out at polymer 100 weight section which has the crosslinkable silyl groups of the example 3 of heat-resistant comparison composition of a hardened material, and it slushed into the 2-mm-thick mold. Vacuum defoaming was carried out and heat cure was carried out for ten days at 50 **. Some obtained hardened material sheets were put into 150 ** oven, it took out 24 hours afterward, and the surface state was observed. It was normal on the surface.
(Example 4 of comparison composition)
205 ml of ethylcyclohexane which the resisting pressure glass polymerization vessels of 2L in which

the polyisobutylene which has an allyl group at the end carried out the synthetic nitrogen purge were made to dry by moleoular sieves and 819 ml of toluene, and 2.89 g (12.5mmol) of p-JIKUMIRU chloride were added. 332 ml (3.91 mol) of isobutylene monomers were introduced into the polymerization vessel, and, in addition to [0.454 g (4.88mmol) of 2-mathylpyridine, and 6.69 ml (61.0mmol) of titanium tetrachlorida] tha next, the polymerization was started. 70 minutes aftar reaction time, 6.86 g (60.0mmol) of allyl trimethylsilane was added, and the introduction reaction of the allyl group was performed to polymer terminals. After washing a reaction solution with water 120 minutes after reaction time, the polyisobutylene which has an allyl group at the end was obtained by distilling off a solvent. After carrying out temperature up of the polymer 200g which has an allyl group at the end obtained by the synthetic above of polyisobutylene which has crosslinkable silyl groups at the end to about 75 **,eq/methyl dimethoxysilane 1.5 [vinyl group] platinum (vinyl siloxane) complex 5×10^{-5} [eq/vinyl group] was added, and the hydrosilylation reaction was performed. FT-IR performed reaction pursuit and the olefin absorption of 1640 cm^{-1} disappeared in about 20 hours. The viscosity of the polyisobutylene which has crosslinkable silyl groups at the obtained end was 360 Pa-s, and number average molecular weights were 4800 and molecular weight distribution 1.52 in GPC measurement (mobile phase chloroform, polystyrene conversion). The number of the crosslinkable silyl groupa per one molecule of polymers for which it asked from ^{1}H-NMR analysia was 1.66 pieces.
(Comparative example 5) Mixed stirring of water 1 weight aection and the dibutyl-tin-dimethoxide 1 weight section was carriad out at polymar 100 weight section which has crosslinkable silyl groupa at the end of the example 4 of heat-resistant comparison compoaition of a hardened material, and it slushed into the 2-mm-thick mold. Vacuum dafoaming was carried out and heat cure waa carried out for ten days at 50 **. Some obtained hardened material shaets were put into 150 ** oven, it took out 24 hours afterward, and the surface state was observed. The surface was dissolving and the liquid object was flowing out in part. The result of Example 8 and the comparative examples 4 and 5 was shown in Table 2.
[0064]
[Table 2]

	実施例8	比較例4	比較例5
重合体	ポリ(アクリル酸n-ブチル)	ポリジメチルシロキサン	ポリイソブチレン
硬化物の耐熱性評価	異常なし	異常なし	表面溶解

[0065]Since the hardened material of tha vinyl system polymer which has the crosslinkabla silyl groups of this invention haa the heat resistance of a silicone series polymar and the level and excels the polyiaobutylane aystam in heat resistance, it can be uaad for tha heat-resistant use demanded.
(Example 9) The acoelerated weatharing teat was done using the sunshine waatharometer for some poly (acrylic acid-n-butyl) hardened material sheets which have the crosslinkable silyl groups obtained in accelerated-weathering-resistance example 5, and the surfaoe state was observed. In the surface dissolution or discoloration, after 1000-hour progress had not happened.
(Comparative examples 6 and 7) Instead of the poly (acrylic acid-n-butyl) hardened material which has the crosslinkable silyl groups obtained in Example 5, In the comparative example 6, the accelerated weathering test was done like Example 9 using the polyisobutylene system polymer which obtained the silicone series polymer obtained in the example 4 of comparison composition in the example 5 of comparison composition by the comparative example 7. As for the comparative example 6, in the surface dissolution or discoloration, after 1000-hour progress had not happened. On the other hand in the comparative example 7, the dissolution of the surface had started after 500-hour progress.
[0066]Since the vinyl system polymer composition which has the crosslinkable silyl groups of this invention has the weatharability of a ailicone series polymer composition and the level and is far superior to the polyiso BURIREN system, it can ba used for the use as which weatherability is required.
(Exampla 10) Azeotropy drying of the polymar 100 weight aection whioh has the crosslinkable ailyl groups obtained in the example 5 of 1 dapth part hardenability composition was carried out with

toluene. Methyl trimetoxysilane 1 weight section and dibutyltin diacetyl acetonato 1 weight section ware addad one by ona under a nitrogan atmosphere, and the 1 liquid compound was produoed by carrying out aealing preservation to the sample bottle. It paid out the aample tube after one-week preservation in the air-conditionad room (23 **60%RH). It waa 3 mm, aa a result of taking out a hardening portion 24 hours after expenditure and measuring the thickness of the depth direction.
(Comparative examples 8 and 9) Instead of the polymar obtainad in tha exampla 5 of 1 dapth part hardenability composition, deptha hardenability was measured like Example 9 using the polyisobutylene system polymer which obtained the silicone series polymer obtained in the example 4 of comparison composition in the example 5 of comparison composition by the comparative example 9 by the comparative example 8. The depths hardenability of the comparative example 8 was 3 mm. In the comparative example 9, the inside was not hardened only by a pellicle stretching on the surface. The vinyl system polymer composition which has the crosslinkable silyl groups of this invention has the 1 depth part hardenability of a silicone series constituent and the level, and since it excels farther than a polyiso BURIREN system constituent, it can be used for it as constituents, such as 1 liquid type sealant.
(Example 11) To poly (acrylic acid-n-butyl) 100 weight section which has the crosslinkable silyl groups obtained in the example 5 of adhesive composition, colloid calcium carbonate 120 weight section and 50-fold dioctyl phthalate — a dragon — the part, the amount part of crosslinkable-silyl-groupa content compound-A-1120 (made by Nippon Unicar) duplexs which has an amino group, and dibutyltin diacetyl acetonato 1 weight saction could ba added, and it mixed, and constructed in the shape of a bead on tha glasa base material. The adhesive property was evaluated by putting in and taaring off slitting to an interface after seven-day neglect at a room tamparature. The destructive situation was the cohesive failura of the combination hardened material. The oonstituent of the vinyl system polymer which has the crosslinkable silyl groups of this invention has aufficient adhesive property, and it can be enough used for it as an existing adhesive hardenability constituent.
(Example 12) To poly (acrylic acid-n-butyl) 100 weight section which has the crosslinkable silyl groups obtained in the example 5 of paintwork composition. The reactant of titanium oxide 10 weight section, colloid calcium carbonate 100 weight section, heavy-calcium-carbonate 40 weight section, octylic acid tin 3 weight section, and lauryl amine 0.75 weight section could be added, it mixed, and the sheet was produced. The acrylic emulsion paint (top of aquosity, Nippon Paint make) diluted with 10% of water was applied on the day following sheet production. It has applied satisfactorily.
(Comparative example 10) The same experiment was conducted using the poly dimethylsiloxane which has the cross-linking silicon group obtained in the example 3 of comparison composition instead of the poly (acrylic acid-n-butyl) which has the crosslinkable silyl groups obtained in the synthetic example 5 in the paintwork exampla 12. Even if it painta, it has crawled immediately. Unlike the constituent which usad the ailicone series polymer, the constituent using the vinyl aystem polymer which has the orosslinkable silyl groupa of thia invention had suffioient paintwork. Therefore, using as hardenability constituanta, such aa sealant whioh oan be painted, is possibla.
(Example 13) To poly (acrylic aoid-n-butyl) 100 weight section which has the crosslinkable silyl groupa obtained in the example 5 of stain resiatance composition. The reactant of titanium oxide 10 weight section, colloid calcium carbonate 100 weight section, heavy-calcium-carbonate 40 weight section, octylic acid tin 3 weight section, and lauryl amine 0.75 weight section could be added, it mixed, the masonry joint of the granite which applied the primer (No.40, Yokohama Rubber make) was filled up, and it was exposed to the outdoors. Even if eight months passed, the circumference of a masonry joint was beautiful. (Comparative example 11) The same experiment was conducted using the poly dimethylsiloxane which has the crosslinkable silyl groups obtained in the example 3 of comparison composition instead of the poly (acrylic acid-n-butyl) which has the crosslinkable silyl groups obtained in the synthetic example 4 in the stain resistance example 13. Progress of eight months would have stained the circumference of the masonry joint darkly. Unlike the constituent which used the silicone series polymer, the constituent using the vinyl system polymer which has the crosslinkable silyl groups of this invention did not have contamination of granite. Therefore, it is possible to use as hardanability conatituents, such as pollution-frea sealant.
(Example 14) To poly (acrylic acid-n-butyl) 100 weight aaction which has the orosslinkable silyl groupa obtained by the same formula as the example 4 of binder composition. 40% toluene solution 175 weight saction (70 weight sections as rosin ester) of spacial rosin ester (the super ester A-100,

product made from the Arakawa chemicals) and the amount part of #918 (tin catalyst, product made from Sankyo organicity) duplexs were mixed, and a 100-micrometer coating machine was used and applied on the PET film. It heated at 50 ** after neglect with the room temperature on the 1st on the 1st. As a result of tearing off 180 degrees and performing adhesive power according to JIS Z 0237, it was 4.5N/25mm. In the vinyl system polymer which has the crosslinkable silyl groups of this invention, it is usable as a binder.
(Synthetic exemple 7) Crosslinkable silyl groups in 200 ml of composition flask of the acrylic acid-n-butyl / methyl methacrylate copolymer which it has The first copper 1.4g (9.8mmol) of bromination, 1.2 g (6.7mmol) of pentamethyl diethylenetriamine, acetonitrile 20mL, Butyl acetate 80mL, 4.4 g (12.2mmol) of 2,5-dibromo diethyl adipate, After teaching 25.0 g (195mmol) of butyl acrylate, 68.4 g (684mmol) of methyl methacrylate, and 5.7 g (24.4mmol) of methyl methacrylete dimethoxy silyl propyl and performing freezing deaeration, 70 ** was made to react under a nitrogen atmosphere for 7 hours. By carrying out removel refining of the copper catelyst through the column of activated alumina, the acrylic acid-n-butyl / methyl methacrylete copolymer which has crosslinkable silyl groups were obtained. The number average molecular weights of the obtained polymer were 12500 and molecular weight distribution 1.55 in GPC measurement (mobile phese chloroform, polystyrene conversion). The viscosity of the 65% toluene solution of the obtained copolymer was 10 Pa-s.
(Example 15) as opposed to solid content 100 weight section of the copolymer obtained in the synthetic example 8 — curing catalyst (#918, product made from Sankyo organicity) 1 weight section of a tin series — in addition, the coat was produced on the steel plate and the Teflon sheet by a 150-micrometer coating machine. The 60-degree mirror reflectivity which measured the coat produced on the steel plate two days after room temperature neglect was 96. It was room temperature 1 50 ** recuperated on the 3rd in the coat produced on the Teflon sheet. [per day] The amount of [which carried out reduced pressure drying of the piece of a coat put into the wire gauze to toluene for 4 hours, and asked it for it at 80 ** after one day immersion] gel was 86%.
(Example 5 of comparison composition) 800 g of synthetic toluene of the acrylic acid-n-butyl / methyl methacrylate copolymer which has crosslinkable silyl groups. It polymerized at 105 ** for 7 hours, carrying out nitrogen bubbling of 208 g of butyl acrylate, 552 g of methyl methacrylate, 40 g of methyl methacrylate dimethoxy silyl propyl, and 24 g of the azobisisobutyronitrile in 2L flask. From GPC measurement (mobile phase chloroform, polystyrene conversion), the number average molecular weight of the acrylic acid-n-butyl / methyl methacrylate copolymer which has the obtained crosslinkable silyl groups was 7400, and molecular weight distribution was 1.87. The viscosity of the 69% toluene solution of the obtained copolymer was 10 Pa-s.
(Comparative example 12) as opposed to solid content 100 weight section of the copolymer obtained in the example 4 of comparison composition like Example 15 — curing catalyst (#918, product made from Sankyo organicity) 1 weight section of a tin series — in addition, the coat was produced on the steel plate and the Teflon sheet by a 150-micrometer coating machine. The 60-degree mirror reflectivity which measured the coat produced on the steel plate two days after room temperature neglect was 96. It was room temparature 1 50 ** recuperated on the 3rd in the coat produced on the Teflon sheet. [per day] The amount of [which carried out reduced pressure drying of the piece of a coat put into the wire gauze to toluene for 4 hours, and asked it for it at 80 ** after one day immersion] gel was 71%.
[0067]Since the vinyl system polymer which has the crosslinkable silyl groups of this invention has narrow molecular weight distribution, while the rise of viscosity is small and high-solid-izing is possible, the paint of the high high gloss for gel can be obtained also with the amount object of polymers.

[Translation done.]